US007232781B2

(12) **United States Patent**
Ishitobi et al.

(10) **Patent No.: US 7,232,781 B2**
(45) **Date of Patent: Jun. 19, 2007**

(54) **DIELECTRIC CERAMIC COMPOSITION, DIELECTRIC CERAMIC, AND LAMINATED CERAMIC PART INCLUDING THE SAME**

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(*) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 111 days.

(21) Appl. No.: **10/543,234**

(22) PCT Filed: **Jan. 20, 2004**

(86) PCT No.: **PCT/JP2004/000423**
§ 371 (c)(1),
(2), (4) Date: **Jul. 22, 2005**

(87) PCT Pub. No.: **WO2004/065325**

PCT Pub. Date: **Aug. 5, 2004**

(65) **Prior Publication Data**

US 2006/0142141 A1 Jun. 29, 2006

(30) **Foreign Application Priority Data**

Jan. 24, 2003	(JP)	2003-016004
Mar. 13, 2003	(JP)	2003-067719
Apr. 3, 2003	(JP)	2003-100676
Jun. 2, 2003	(JP)	2003-156949

(51) **Int. Cl.**

C04B 35/465	(2006.01)
B32B 18/00	(2006.01)
B32B 17/06	(2006.01)

(52) **U.S. Cl.** **501/136**; 501/122; 428/210; 428/701; 428/702

(58) **Field of Classification Search** 501/136, 501/122; 428/210, 701, 702
See application file for complete search history.

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(57) **ABSTRACT**

A dielectric ceramic composition which can be sintered at such a temperature of about 800 to 1000° C. as to permit incorporation of and multilayer formation with a low resistant conductor such as Ag or Cu by the simultaneous sintering with the low resistant conductor, which is sintered to form dielectric ceramics having a dielectric constant ϵ_r of not more than 10, and a resonator having a large $Q \times f_0$ value and an absolute value in temperature coefficient τ_f of resonance frequency f_0 of not more than 20 ppm/° C., the value being easy to be controlled. The dielectric ceramic composition contains a glass component in an amount of 5 to 150 parts by weight based on 100 parts by weight of a main component represented by general formula: $aZnAl_2O_4$-bZn_2SiO_4-$cTiO_2$-dZn_2TiO_4, in which the molar fractions of respective components a, b, c, and d satisfy $5.0 \leqq a \leqq 80.0$ mol % $5.0 \leqq b \leqq 70.0$ mol %, $5.0 \leqq c \leqq 27.5$ mol %, $0 \leqq d \leqq 30.0$ mol % (a+b+c+d=100 mol %) The main component may also be represented by aMg_2SiO_4 -bZn2Al_2O_4 -$cSiO_2$ -dCaTiO_3 -eZn_2SiO_4.

7 Claims, 7 Drawing Sheets

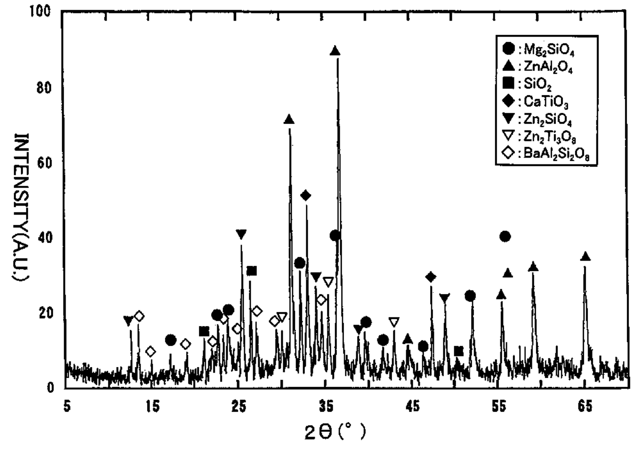


FIG. 1

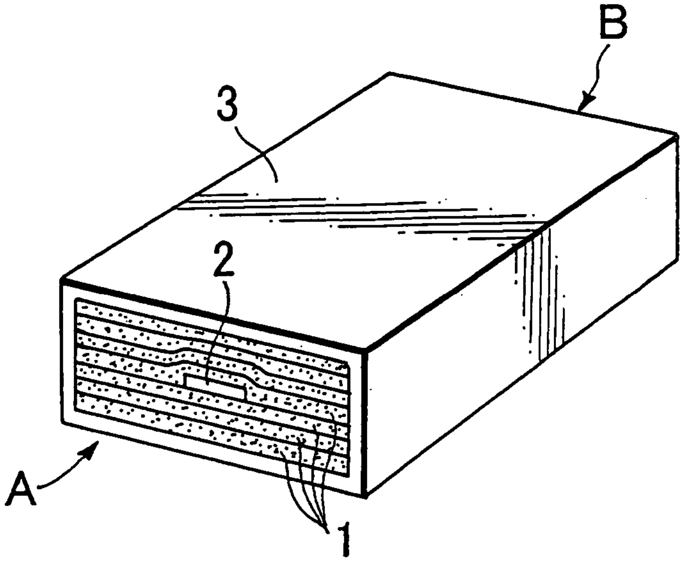


FIG. 2

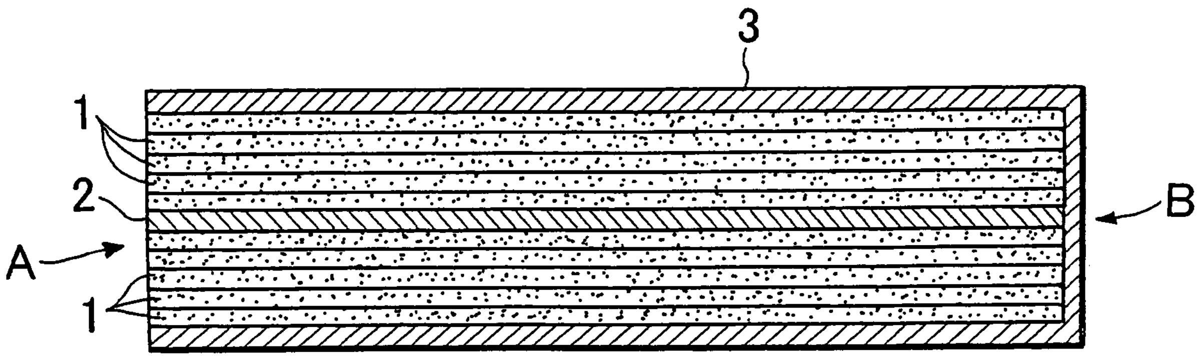


FIG.3

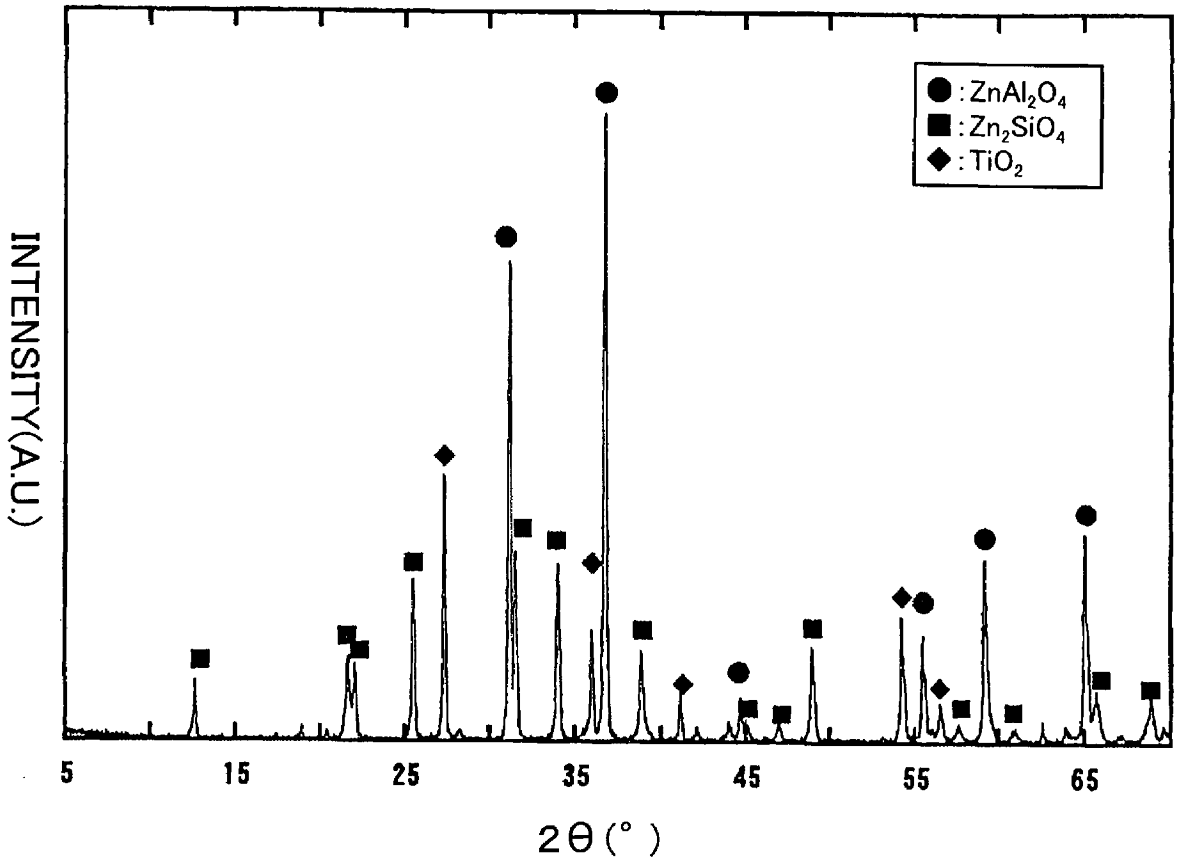
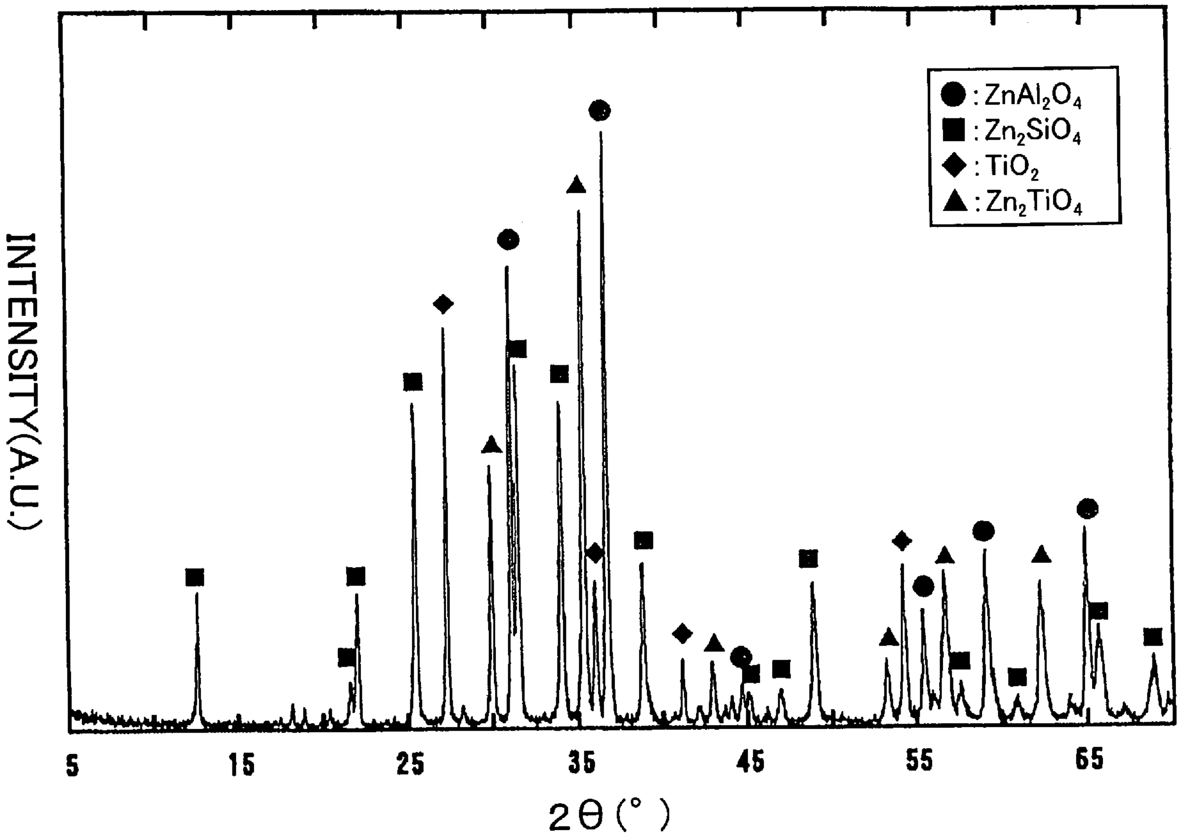
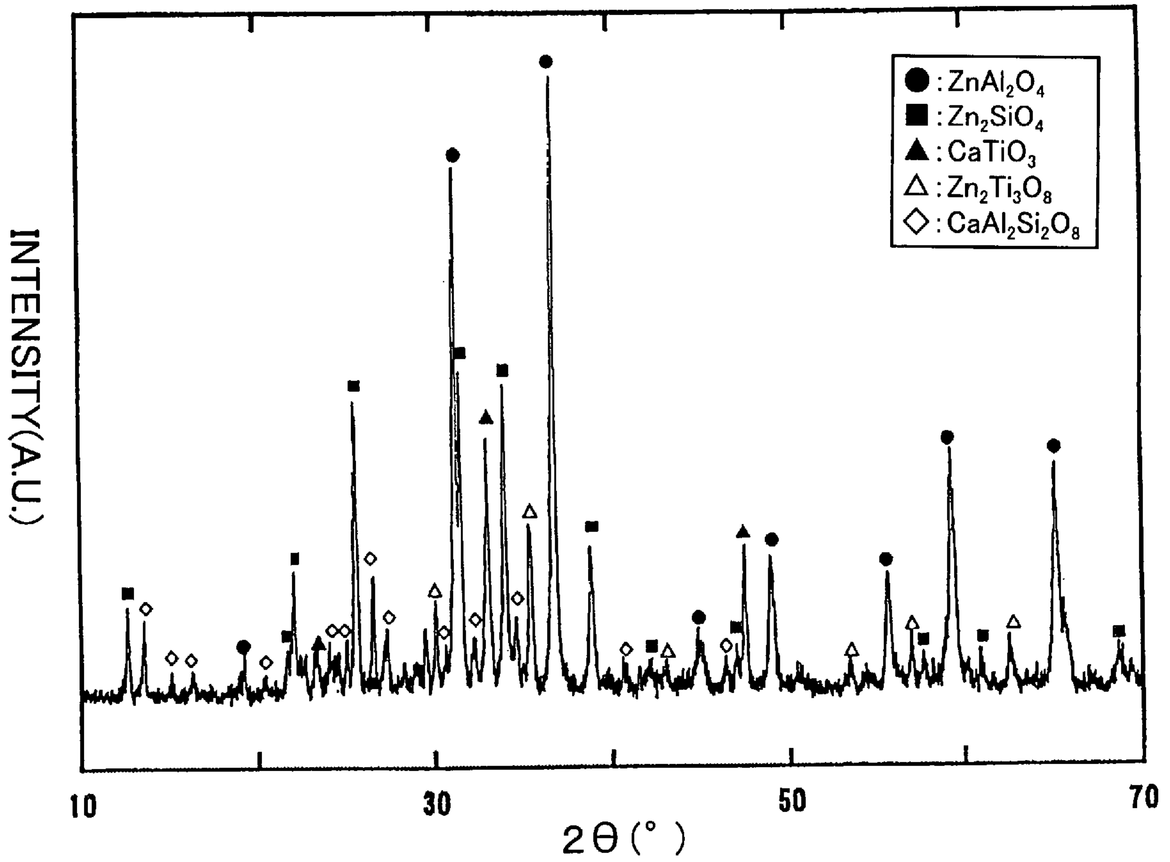


FIG.4



● : $ZnAl_2O_4$
■ : Zn_2SiO_4
◆ : TiO_2
▲ : Zn_2TiO_4
INTENSITY(A.U.)
5
15
25
35
45
55
65
2θ(°)

FIG.5



● : $ZnAl_2O_4$
■ : Zn_2SiO_4
▲ : $CaTiO_3$
△ : $Zn_2Ti_3O_8$
◇ : $CaAl_2Si_2O_8$
INTENSITY(A.U.)
10
30
50
70
2θ(°)

FIG.6

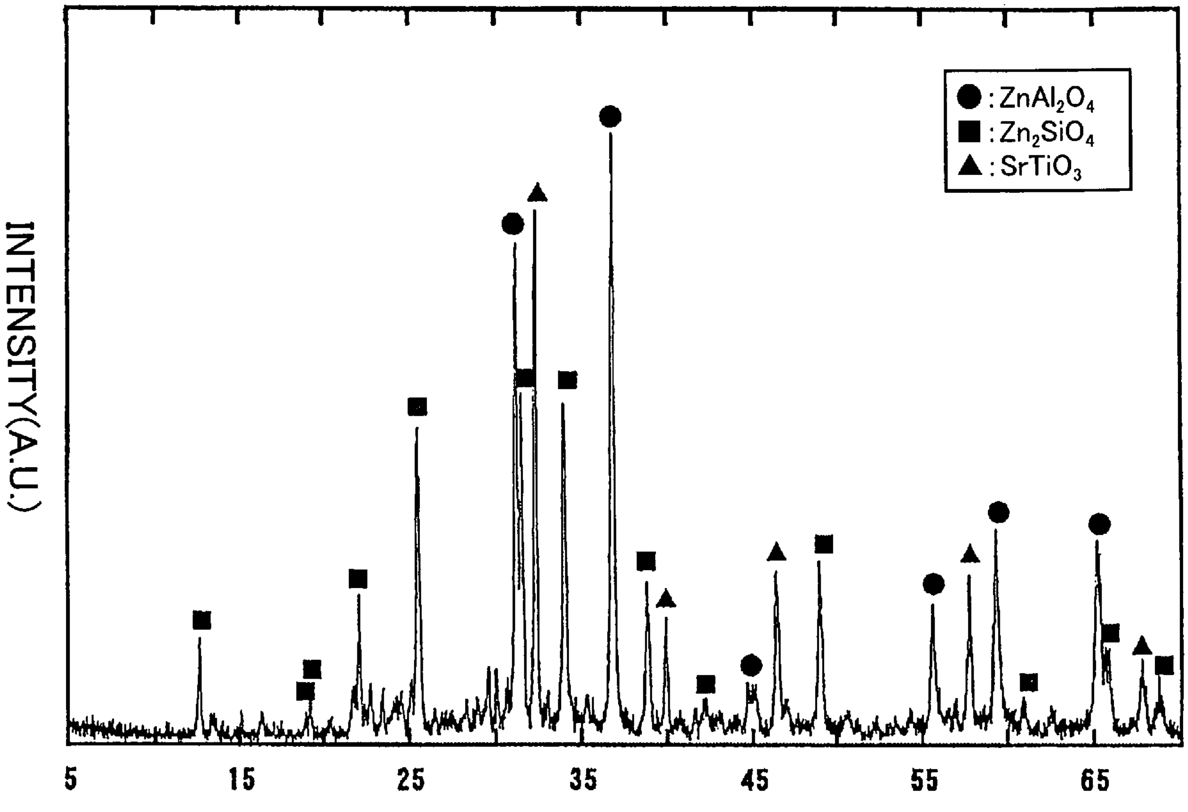


FIG.7

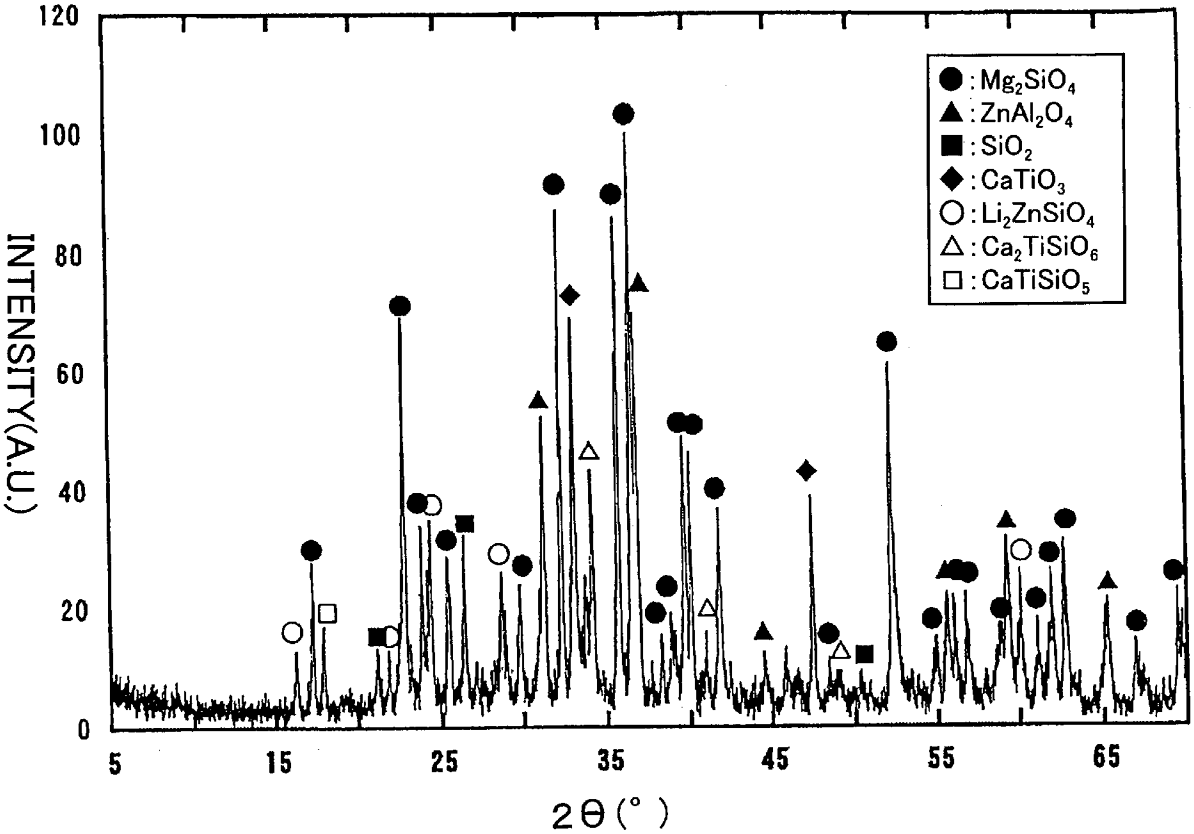
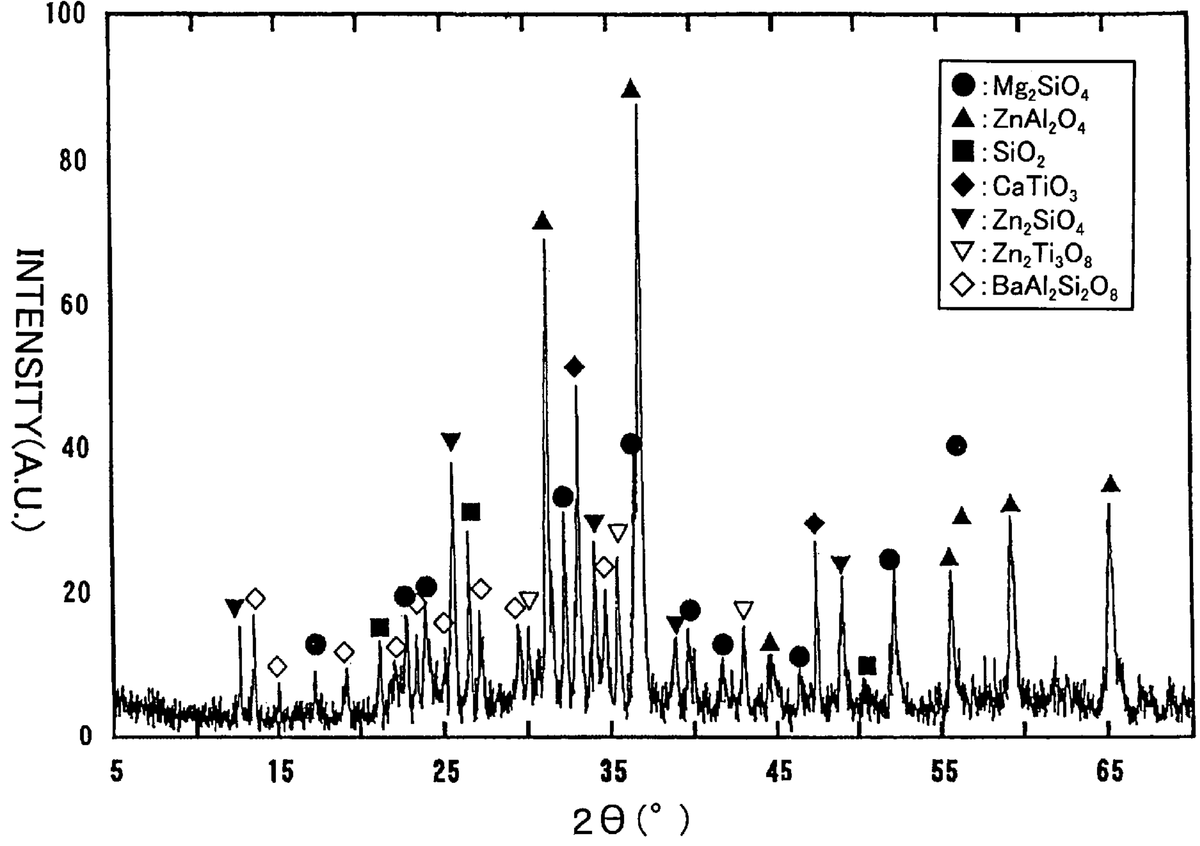

120
100
80
60
40
20
0
INTENSITY(A.U.)
5
15
25
35
45
55
65
2θ(°)
●: Mg_2SiO_4
▲: $ZnAl_2O_4$
■: SiO_2
◆: $CaTiO_3$
○: Li_2ZnSiO_4
△: Ca_2TiSiO_6
□: $CaTiSiO_5$

FIG.8



100
80
60
40
20
0
INTENSITY(A.U.)
5
15
25
35
45
55
65
2θ(°)
●: Mg_2SiO_4
▲: $ZnAl_2O_4$
■: SiO_2
◆: $CaTiO_3$
▼: Zn_2SiO_4
▽: $Zn_2Ti_3O_8$
◇: $BaAl_2Si_2O_8$

DIELECTRIC CERAMIC COMPOSITION, DIELECTRIC CERAMIC, AND LAMINATED CERAMIC PART INCLUDING THE SAME

This application is a 371 of PCT/JP2004/000423 filed on Jan. 20, 2004, published on Aug. 5, 2004 under publication number WO 2004/065325 A1 which claims priority benefits from Japanese Patent Application Number 2003-016004 filed Jan. 24, 2003 and Japanese Patent Application Number 2003-067719 filed Mar. 13, 2003 and Japanese Patent Application Number 2003-100676 filed Apr. 3, 2003 and Japanese Patent Application Number 2003-156949 filed Jun. 2, 2003.

TECHNICAL FIELD

The present invention relates to a dielectric ceramic composition that has a dielectric constant ϵ_r of not more than 10, a large Q-value in high-frequency region such as microwave or millimeter wave, and a small absolute value in temperature coefficient τ_f of resonance frequency f_0 and that can be sintered simultaneously with Ag, Cu or the like as a low resistant conductor, dielectric ceramics obtained by sintering the dielectric ceramic composition, and a laminated ceramic part using the dielectric ceramics, such as a laminated dielectric resonator, a laminated dielectric filter, and a laminated dielectric substrate.

BACKGROUND ART

In recent years, along with a rapid development of communication networks, frequency range to be used for the communication is extended to cover high-frequency region such as microwave region or millimeter wave region. With regards to the dielectric ceramic composition for high frequency, it is demanded that a dielectric resonator using dielectric ceramics obtained by sintering the dielectric ceramic composition has a large unloaded Q-value. Further, the dielectric ceramic composition for high frequency is demanded to have a small absolute value in temperature coefficient τ_f of resonance frequency f_0. On the other hand, as the dielectric constant ϵ_r of the dielectric ceramics is larger, a microwave circuit or millimeter wave circuit can be more reduced in the size. However, in terms of high-frequency region corresponding to microwave region, when the dielectric constant ϵ_r becomes too large, the circuit is excessively reduced in the size, with the result that high processing precision is demanded. Therefore, a material having a small dielectric constant ϵ_r is required.

As the dielectric ceramic composition for manufacturing a dielectric resonator having a large Q-value and a small absolute value in temperature coefficient τ_f of the resonance frequency f_0, $BaO—MgO—WO_3$-base material (refer to JP(A)-6-236708 (paragraph number [0033] on page 11, tables 1 to 8)), $MgTiO_3—CaTiO_3$-base material (refer to JP(A)-6-199568 (paragraph number [0018] on page 5, tables 1 to 3)), and the like have been proposed. However, the dielectric constant ϵ_r of the dielectric ceramics obtained from the above ceramic compositions exceeds 10. Hence, the dielectric ceramic composition from which dielectric ceramics having a lower dielectric constant can be manufactured is demanded.

Forsterite (Mg_2SiO_4) and Alumina (Al_2O_3), which have relatively small dielectric constants ϵ_r of 7 and 10 respectively, are known as the dielectric ceramic composition from which a dielectric ceramics excellent in high-frequency characteristics can be manufactured. However, the temperature dependency (τ_f) of resonance frequency is large on the minus side (−60 ppm/° C.), so that an application to such uses as the dielectric resonator and dielectric filter where the temperature dependency needs to be small is limited.

In recent years, laminated ceramic parts formed by laminating dielectric ceramics, such as a laminated dielectric resonator, a laminated dielectric filter, or a laminated dielectric substrate have been developed and the lamination by the simultaneous sintering of a dielectric ceramic composition and an internal electrode is being performed. However, the above-described dielectric ceramic compositions have a difficulty in performing the simultaneous sintering with the internal electrode because of their high sintering temperature of 1300° C. or more and therefore, for forming a lamination structure, material of the internal electrode is limited to an expensive high-temperature resistant material such as platinum (Pt). For this reason, there has been demanded a dielectric ceramic composition capable of performing the simultaneous sintering with the internal electrode at a low temperature of 1000° C. or less, using as the internal electrode material silver (Ag), Ag—Pd, Cu and the like, which are low resistant and inexpensive conductors.

As the dielectric ceramics having a small dielectric constant and capable of performing the sintering at a low temperature, ceramics comprising a $ZnAl_2O_4$ crystal, an α-SiO_2 crystal, a Zn_2SiO_4 crystal, and a glass phase is known (refer to JP(A)-2002-338341 (paragraph number [0050] on page 10, table 4, etc.)). This material is a printed circuit board material including the glass phase and, therefore, a mechanical strength is stressed in it. However, the Q-value of the resonator is not sufficient for a high-frequency dielectric ceramics. Further, there is no description about the temperature coefficient τ_f of resonance frequency f_0 in the above publication.

In addition, as the dielectric ceramics having a small dielectric constant and capable of performing the sintering at a low temperature, a ceramics comprising SiO_2, Al_2O_3, MgO, ZnO, and B_2O_3, where crystalline phases of ZnO and Al_2O_3, crystalline phases of ZnO and SiO_2, crystalline phases of MgO and SiO_2, and amorphous phase of SiO_2 or amorphous phases of SiO_2 and B_2O_3 are present together is known (refer to JP(A)-2002-53368 (paragraph number [0053] on page 5, table 2, etc.)). This material is a printed circuit board material including the glass phase and, therefore, a mechanical strength is stressed in it. However, the Q-value of the resonator is not sufficient for high-frequency dielectric ceramics. Further, there is no description about the temperature coefficient τ_f of resonance frequency f_0 in the above publication.

DISCLOSURE OF THE INVENTION

An object of the present invention is to provide a dielectric ceramic composition which is capable of solving the above problem, which can be sintered at such a temperature of about 800 to 1000° C. as to permit incorporation of and multilayer formation with a low resistant conductor such as Ag or Cu by the simultaneous sintering with the low resistant conductor, which is sintered to form dielectric ceramics having a dielectric constant ϵ_r of not more than 10, and a resonator having a large $Q \times f_0$ value and an absolute value in temperature coefficient τ_f of resonance frequency f_0 of not more than 20 ppm/° C., the value being easy to be controlled. Another object of the present invention is to provide a laminated ceramic part such as a laminated dielectric resonator, a laminated filter, or a laminated dielectric substrate, which has dielectric layers obtained by sintering the above dielectric ceramic composition and an internal electrode mainly comprising Ag or Cu.

To achieve the above object, according to a first aspect of the present invention, there is provided a dielectric ceramic composition containing a glass component in an amount of 5 to 150 parts by weight based on 100 parts by weight of a main component represented by general formula (1): $aZnAl_2O_4$-bZn_2SiO_4-$cTiO_2$-dZn_2TiO_4, in which the molar fractions of respective components a, b, c, and d satisfy $5.0 \leqq a \leqq 80.0$ mol %, $5.0 \leqq b \leqq 70.0$ mol %, $5.0 \leqq c \leqq 27.5$ mol %, $0 \leqq d \leqq 30.0$ mol % (a+b+c+d=100 mol %).

In the first aspect of the present invention, the glass component may include one or more glasses selected from a PbO-base glass, a ZnO-base glass, a SiO_2-base glass, a B_2O_3-base glass, and a glass comprising two or more oxides selected from the group consisting of SiO_2, Al_2O_3, ZnO, PbO, Bi_2O_3, BaO, SrO, CaO, SnO_2, ZrO_2, and B_2O_3.

By sintering the above dielectric ceramic composition, dielectric ceramics containing crystalline phases of $ZnAl_2O_4$, Zn_2SiO_4, and Zn_2TiO_4 and a glass phase, or containing crystalline phases of $ZnAl_2O_4$, Zn_2SiO_4, TiO_2 and Zn_2TiO_4 and a glass phase are obtained.

To achieve the above object, according to a second aspect of the present invention, there is provided a dielectric ceramic composition containing a Li compound as a subcomponent in an amount of 2 to 30 parts by weight in terms of Li_2O and a glass component in an amount of 5 to 150 parts by weight, based on 100 parts by weight of a main component comprising a calcined body obtained by calcining a material composition represented by general formula (2): $aZnO$-bAl_2O_3-$cSiO_2$-$d(xCaO$-$(1-x)TiO_2)$, in which the molar fractions of respective components a, b, c, and d satisfy $7.5 \leqq a \leqq 55.0$ mol %, $5.0 \leqq b \leqq 65.0$ mol %, $5.0 \leqq c \leqq 70.0$ mol %, $7.5 \leqq d \leqq 27.5$ mol % (a+b+c+d=100 mol %) and x satisfies $0 \leqq x \leqq 0.75$.

In the second aspect of the present invention, the main component may contain a $ZnAl_2O_4$ crystal, a Zn_2SiO_4 crystal, and at least one of a $CaTiO_3$ crystal and a TiO_2 crystal. In the second aspect of the present invention, the glass component may include one or more glasses selected from a PbO-base glass, a ZnO-base glass, a SiO_2-base glass, a B_2O_3-base glass, and a glass comprising two or more oxides selected from the group consisting of SiO_2, Al_2O_3, ZnO, PbO, Bi_2O_3, BaO, SrO, SnO_2, ZrO_2, and B_2O_3. Further, in the second aspect of the present invention, the glass component may be composed of SiO_2 in an amount of 2.5 to 70 wt %, Al_2O_3 in an amount of 0 to 15 wt %, ZnO in an amount of 10 to 55 wt %, PbO in an amount of 0 to 35 wt %, Bi_2O_3 in an amount of 0 to 2 wt %, BaO in an amount of 0 to 5 wt %, SrO in an amount of 0 to 2 wt %, SnO_2 in an amount of 0 to 2 wt %, ZrO_2 in an amount of 0 to 1 wt %, and B_2O_3 in an amount of 10 to 50 wt %.

By sintering the above dielectric ceramic composition, dielectric ceramics containing one or more crystalline phases of $ZnAl_2O_4$, Zn_2SiO_4, and at least one of $CaTiO_3$ and TiO_2 and a glass phase are obtained.

The above dielectric ceramic composition can be produced by mixing a Li compound as a subcomponent in an amount of 2 to 30 parts by weight in terms of Li_2O and a glass component in an amount of 5 to 150 parts by weight, based on 100 parts by weight of a main component comprising a calcined body obtained by calcining, at from 900 to 1200° C., a material composition represented by general formula (2), in which the molar fractions of respective components a, b, c, and d and coefficient x fall within the above ranges.

To achieve the above object, according to a third aspect of the present invention, there is provided a dielectric ceramic composition containing a Li compound as a subcomponent in an amount of 2 to 30 parts by weight in terms of Li_2O and a glass component in an amount of 5 to 150 parts by weight, based on 100 parts by weight of a main component represented by general formula (3): $aZnAl_2O_4$-bZn_2SiO_4-$cSiO_2$-$dSrTiO_3$, in which the molar fractions of respective components a, b, c, and d satisfy $2.5 \leqq a \leqq 77.5$ mol %, $2.5 \leqq b \leqq 77.5$ mol %, $2.5 \leqq c \leqq 37.5$ mol %, $10.0 \leqq d \leqq 17.5$ mol % (a+b+c+d=100 mol %).

In the third aspect of the present invention, the glass component may include one or more glasses selected from a PbO-base glass, a ZnO-base glass, a SiO_2-base glass, a B_2O_3-base glass, and a glass comprising two or more oxides selected from the group consisting of SiO_2, Al_2O_3, ZnO, PbO, Bi_2O_3, BaO, SrO, SnO_2, ZrO_2, and B_2O_3. Further, in the third aspect of the present invention, the glass component may be composed of SiO_2 in an amount of 2.5 to 70 wt %, Al_2O_3 in an amount of 0 to 15 wt %, ZnO in an amount of 10 to 55 wt %, PbO in an amount of 0 to 35 wt %, Bi_2O_3 in an amount of 0 to 2 wt %, BaO in an amount of 0 to 5 wt %, SrO in an amount of 0 to 2 wt %, SnO_2 in an amount of 0 to 2 wt %, ZrO_2 in an amount of 0 to 1 wt %, and B_2O_3 in an amount of 10 to 50 wt %.

By sintering the above dielectric ceramic composition, dielectric ceramics containing crystalline phases of $ZnAl_2O_4$, Zn_2SiO_4, and $SrTiO_3$ and a glass phase are obtained.

To achieve the above object, according to a fourth aspect of the present invention, there is provided a dielectric ceramic composition containing a Li compound as a subcomponent in an amount of 1 to 15 parts by weight in terms of Li_2O and a glass component in an amount of 5 to 150 parts by weight, based on 100 parts by weight of a main component represented by general formula (4): aMg_2SiO_4-$bZnAl_2O_4$-$cSiO_2$-$dCaTiO_3$-eZn_2SiO_4, in which the molar fractions of respective components a, b, c, d, and e satisfy $0.10 \leqq a \leqq 0.72$, $0.08 \leqq b \leqq 0.62$, $0.02 \leqq c \leqq 0.22$, $0.12 \leqq d \leqq 0.22$, $0 \leqq e \leqq 0.08$ (a+b+c+d+e=1).

In the fourth aspect of the present invention, the glass component may include one or more glasses selected from a PbO-base glass, a ZnO-base glass, a SiO_2-base glass, a B_2O_3-base glass, and a glass comprising two or more oxides selected from the group consisting of SiO_2, Al_2O_3, ZnO, PbO, Bi_2O_3, BaO, SrO, SnO_2, ZrO_2, and B_2O_3. Further, in the fourth aspect of the present invention, the glass component may be composed of SiO_2 in an amount of 2 to 70 wt %, Al_2O_3 in an amount of 0 to 15 wt %, ZnO in an amount of 10 to 55 wt %, PbO in an amount of 0 to 35 wt %, Bi_2O_3 in an amount of 0 to 2 wt %, BaO in an amount of 0 to 30 wt %, SrO in an amount of 0 to 2 wt %, SnO_2 in an amount of 0 to 2 wt %, ZrO_2 in an amount of 0 to 1 wt %, and B_2O_3 in an amount of 10 to 50 wt %.

By sintering the above dielectric ceramic composition, dielectric ceramics containing crystalline phases of Mg_2SiO_4, $ZnAl_2O_4$, SiO_2, and $CaTiO_3$, and a glass phase, or containing crystalline phases of Mg_2SiO_4, $ZnAl_2O_4$, SiO_2, $CaTiO_3$, and Zn_2SiO_4, and a glass phase are obtained.

Further, according to the present invention, there is provided a dielectric ceramic part having a plurality of dielectric layers, an internal electrode formed between the dielectric layers and an external electrode electrically connected to the internal electrode, characterized in that the dielectric layers are constituted of dielectric ceramics obtained by sintering: the dielectric ceramic composition comprising a main component represented by the general formula (1); the dielectric ceramic composition comprising a calcined body of a material composition represented by the general formula (2) as a main component; the dielectric ceramic composition comprising a main component represented by the general formula (3); or the dielectric ceramic composition comprising a main component represented by the general formula (4), and the internal electrode is made of elemental Cu or elemental Ag, or an alloy material mainly comprising Cu or Ag.

Since the dielectric ceramic composition according to the present invention can be sintered at a sintering temperature of 1000° C. or less, it is possible to perform simultaneous sintering with a low resistant conductor such as Cu or Ag. Further, by sintering the dielectric ceramic composition according to the present invention, it is possible to provide ceramics exhibiting a large $Q\times f_0$ value, which is a product of resonance frequency f_0 (GHz) and Q-value, of 10000 (GHz) or more, or in some cases, 20000 (GHz) or more, and having a low dielectric loss. Further, the dielectric ceramic composition according to the present invention can provide ceramics having an absolute value in temperature coefficient (τ_f) of resonance frequency of not more than 20 ppm/° C., and thereby less influenced by temperature. Further, dielectric constant ϵ_r of dielectric ceramics obtained from the dielectric ceramic composition according to the present invention is not more than 10, so that a high-frequency device or high-frequency circuit obtained using the dielectric ceramics is not excessively reduced in the size, but can be kept in an appropriate size, resulting in excellent processing accuracy and productivity.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. **1** is a perspective view showing an embodiment of a laminated ceramic part according to the present invention;

FIG. **2** is a cross-sectional view of the laminated ceramic part of FIG. **1**;

FIG. **3** is an X-ray diffraction pattern of dielectric ceramics obtained in example 2;

FIG. **4** is an X-ray diffraction pattern of dielectric ceramics obtained in example 13;

FIG. **5** is an X-ray diffraction pattern of dielectric ceramics obtained in example 33;

FIG. **6** is an X-ray diffraction pattern of dielectric ceramics obtained in example 44;

FIG. **7** is an X-ray diffraction pattern of dielectric ceramics obtained in example 68; and

FIG. **8** is an X-ray diffraction pattern of dielectric ceramics obtained in example 76.

BEST MODE FOR CARRYING OUT THE INVENTION

Hereinafter, embodiments of the present invention will be described.

(1) FIRST EMBODIMENT

(Embodiment Related to a Dielectric Ceramic Composition Comprising a Composition Represented by the above General Formula (1) as a Main Component)

A dielectric ceramic composition according to the present embodiment contains a glass component in an amount of 5 to 150 parts by weight based on 100 parts by weight of the main component represented by the general formula (1): $a\mathrm{ZnAl_2O_4}$-$b\mathrm{Zn_2SiO_4}$-$c\mathrm{TiO_2}$-$d\mathrm{Zn_2TiO_4}$.

The glass component to be used is in the form of a glass or powdered glass (glass powder). The glass used herein indicates an amorphous solid substance and can be obtained by fusion. The powdered glass or glass powder is obtained by pulverizing a glass. A crystallized glass partially containing a crystallized substance therein is also included in the glass. Hereinafter, the glass that is referred to as merely "glass" includes the powdered glass or crystallized glass. This is the same for all the following embodiments.

The glass component for use in the present embodiment includes a PbO-base glass, a ZnO-base glass, a SiO_2-base glass, a B_2O_3-base glass, and a glass comprising metal oxides. The PbO-base glass is a glass containing PbO, and examples thereof include a glass containing PbO—SiO_2, PbO—B_2O_3 or PbO—P_2O_5, or a glass containing R_2O—PbO—SiO_2, R_2O—CaO—PbO—SiO_2, R_2O—ZnO—PbO—SiO_2 or R_2O—Al_2O_3—PbO—SiO_2 (herein, R_2O is Na_2O or K_2O (this is the same for all the following embodiments)). The ZnO-base glass is a glass containing ZnO, and examples thereof include a glass containing ZnO—Al_2O_3—BaO—SiO_2 or ZnO—Al_2O_3—R_2O—SiO_2. The SiO_2-base glass is a glass containing SiO_2, and examples thereof include a glass containing SiO_2—Al_2O_3—R_2O or SiO_2—Al_2O_3—BaO. The B_2O_3-base glass is a glass containing B_2O_3, and examples thereof include a glass containing B_2O_3—SiO_2—ZnO, or B_2O_3—Al_2O_3—R_2O.

As the glass component for use in the present embodiment, in addition to the PbO-base glass, the ZnO-base glass, the SiO_2-base glass, and the B_2O_3 glass, a glass comprising various metal oxides can also be used, and examples thereof include a glass comprising two or more oxides selected from the group consisting of SiO_2, Al_2O_3, ZnO, PbO, Bi_2O_3, BaO, SrO, CaO, SnO_2, ZrO_2, and B_2O_3. Either an amorphous glass or a crystalline glass may be used as the glass. When the glass contains PbO, the sintering temperature is liable to lower, however, the unloaded Q-value is liable to decrease and therefore, the content of the PbO component in the glass is preferably 40% by weight or less. A glass containing a ZnO component, an Al_2O_3 component, a BaO component, a SiO_2 component, and a B_2O_3 component is more preferably used as the glass for use in the present embodiment in that a high unloaded Q-value can be obtained.

Reasons for limiting the composition in the present embodiment will next be described. If the glass component is contained in an amount of less than 5 parts by weight based on 100 parts by weight of the main component served as the base material of the ceramics obtained by sintering, a preferable sintered body cannot be obtained at 1000° C. or less; whereas if the glass component is contained in excess of 150 parts by weight, the glass is liable to elute in sintering, with the result that a preferable sintered body cannot be obtained.

It is unfavorable that the molar fraction a in the main component is less than 5.0 mol %, or it exceeds 80.0 mol %. In the former case, the $Q\times f_0$ value becomes less than 10000 (GHz); in the latter case, the composition cannot be sintered at 1000° C. or less. Further, it is unfavorable that the molar fraction b in the main component is less than 5.0 mol %, or it exceeds 70.0 mol %. In the former case, a preferable sintered body cannot be obtained; in the latter case, the absolute value in temperature coefficient (τ_f) of resonance frequency becomes more than 20 ppm/° C. Further, it is unfavorable that the molar fraction c in the main component is less than 5.0 mol %, or it exceeds 27.5 mol %. In this case, the absolute value in temperature coefficient (of) of resonance frequency becomes more than 20 ppm/° C. Further, it is unfavorable that the molar fraction d in the main component exceeds 30.0 mol %. In this case, the $Q\times f_0$ value becomes smaller. The dielectric ceramic composition according to the present embodiment may contain other components in addition to the main component thereof as far as the object of the present invention is not impaired.

When the molar fraction d in the main component is 0 mol %, the main component of the dielectric ceramic composition according to the present embodiment is represented by the formula: $aZnAl_2O_4$-bZn_2SiO_4-$cTiO_2$ (a+b+c=100 mol %). The dielectric ceramic composition containing a glass component in an amount of 5 to 150 parts by weight based on 100 parts by weight of the three-phase main component can also obtain the effect of the present invention.

The dielectric ceramic composition comprising a main component containing crystals of four components represented by $aZnAl_2O_4$-bZn_2SiO_4-$cTiO_2$-dZn_2TiO_4 and a glass component can be taken as the most preferable form in the present embodiment. The addition of the Zn_2TiO_4 to the dielectric ceramic composition comprising crystals of the three-phase main component represented by $aZnAl_2O_4$-bZn_2SiO_4-$cTiO_2$ and glass component can lower the sintering temperature and, in particular, makes it harder for a defect such as a migration to occur in the case of simultaneous sintering with Ag which is low melting metal.

A preferable production method of the dielectric ceramic composition according to the present embodiment and dielectric ceramics obtained by sintering the same will next be described. The respective base materials constituting the main component are obtained as follows. $ZnAl_2O_4$ is obtained by mixing ZnO and Al_2O_3 in a molar ratio of 1:1 followed by calcination. Zn_2SiO_4 is obtained by mixing ZnO and SiO_2 in a molar ratio of 2:1 followed by calcination. Zn_2TiO_4 is obtained by mixing ZnO and TiO_2 in a molar ratio of 2:1 followed by calcination. Predetermined amounts of the required base materials of the above $ZnAl_2O_4$, Zn_2SiO_4, TiO_2 and Zn_2TiO_4 and glass powder are wet-mixed together with a solvent such as water or an alcohol. Subsequently, after removing the water, the alcohol or the like, an organic binder such as polyvinyl alcohol and water are mixed in the resulting powder. The mixture is rendered uniform, dried and pulverized, followed by molding under pressure (pressure: on the order of from 100 to 1000 kg/cm^2). The molded product obtained is sintered at from 825 to 925° C. in an oxygen-containing gas atmosphere such as air atmosphere, whereby the dielectric ceramics represented by the above composition formula can be obtained.

Further, another example of the preferable production method of the dielectric ceramic composition according to the present embodiment and dielectric ceramics obtained by sintering the same will next be described. Predetermined amounts of the required starting materials of respective powders: zinc oxide (ZnO), aluminium oxide (Al_2O_3), silicon oxide (SiO_2), and titanium oxide (TiO_2) are wet-mixed together with a solvent such as water or an alcohol. Subsequently, after removing the water, the alcohol, or the like, the mixture is calcined at from 800 to 1200° C. for 2 hours in air atmosphere to obtain a calcined powder comprising $ZnAl_2O_4$, Zn_2SiO_4, TiO_2, and Zn_2TiO_4. A predetermined amount of the glass powder is added to thus-obtained calcined powder followed by wet-mixing together with a solvent such as water or an alcohol. Subsequently, after removing the water, the alcohol, or the like, an organic binder such as polyvinyl alcohol and water are mixed in the resulting powder. The mixture is rendered uniform, dried and pulverized, followed by molding under pressure (pressure: on the order of from 100 to 1000 kg/cm^2). The molded product obtained is sintered at from 825 to 925° C. in an oxygen-containing gas atmosphere such as air atmosphere, whereby the dielectric ceramics represented by the above composition formula can be obtained. As materials of zinc, aluminium, silicon, and titanium, carbonate, hydroxide, and an organic metal compound each turning into an oxide at the time of calcining, may also be used, in addition to ZnO, Al_2O_3, SiO_2, and TiO_2.

FIG. **3** shows an X-ray diffraction pattern of the dielectric ceramics according to the present embodiment comprising crystals of the three-phase main component represented by $aZnAl_2O_4$-bZn_2SiO_4-$cTiO_2$ and glass phase, which is obtained by the former production method. FIG. **4** shows an X-ray diffraction pattern of the dielectric ceramics according to the present embodiment comprising crystals of the four-phase main component represented by $aZnAl_2O_4$-bZn_2SiO_4-$cTiO_2$-dZn_2TiO_4 and glass phase, which is obtained by the former production method. Note that it is possible to obtain the dielectric ceramics having the target crystalline structure by using the latter production method in which the oxides are used as starting materials, as well as by using the production method in which carbonate, hydroxide, and an organic metal compound each turning into an oxide at the time of calcining are used as the starting materials.

The dielectric ceramic composition according to the present embodiment is utilized for manufacturing a dielectric resonator. More specifically, the dielectric ceramic composition is processed in an appropriate shape and size, and sintered, followed by formation of required electrodes. Further, the dielectric ceramic composition according to the present embodiment is utilized to obtain various types of laminated ceramic parts. More specifically, resin such as polyvinyl butyral, a plasticizer such as dibutylphthalate, organic solvent such as toluene are mixed in the dielectric ceramic composition, followed by sheet forming using a doctor blade method. The obtained sheet and a conductor are laminated and sintered in an integrated manner. Examples of the laminated dielectric parts include a laminated dielectric resonator, a laminated ceramic condenser, an LC filter, and a dielectric substrate.

The laminated ceramic part according to the present embodiment comprises a plurality of dielectric layers, an internal electrode formed between the dielectric layers and an external electrode electrically connected to the internal electrode. The dielectric layers are constituted of dielectric ceramics obtained by sintering the dielectric ceramic composition and the internal electrode is made of elemental Cu or elemental Ag, or an alloy material mainly comprising Cu or Ag. The laminated ceramic parts according to the present embodiment can be obtained by simultaneously sintering the dielectric layers each containing the dielectric ceramic composition and elemental Cu, elemental Ag or an alloy material mainly comprising Cu or Ag.

Examples of one embodiment of the laminated ceramic parts include a tri-plate type resonator shown in FIG. **1**. FIG. **1** is a perspective view showing the tri-plate type resonator according to the present embodiment, and FIG. **2** is a cross-sectional view of the resonator. As shown in FIGS. **1** and **2**, the tri-plate type resonator is a laminated ceramic part comprising a plurality of dielectric layers **1**, an internal electrode **2** formed between the dielectric layers and an external electrode **3** electrically connected to the internal electrode. The internal electrode **2** is disposed at the center of the laminated dielectric layers **1**. The internal electrode **2** is formed so as to pass through the resonator from a first face A to a second face B opposing the first face A. Only the first face A is an open face. Five faces of the resonator exclusive of the first face A are covered by an external electrode **3**, and the internal electrode **2** and the external electrode **3** are connected to each other on the second face B. The material of the internal electrode **2** contains Cu or Ag, or an alloy material mainly comprising Cu or Ag. When the dielectric ceramic composition according to the present embodiment is used, the sintering can be performed at a low temperature and therefore, these materials for the internal electrode can be used to perform simultaneous sintering.

(2) SECOND EMBODIMENT

(Embodiment Related to a Dielectric Ceramic Composition Comprising a Calcined Body of a Material Composition Represented by the above General Formula (2) as a Main Component)

A dielectric ceramic composition according to the present embodiment comprises a calcined body obtained by calcining a material composition represented by the general formula (2): $a\text{ZnO-}b\text{Al}_2\text{O}_3\text{-}c\text{SiO}_2\text{-}d(x\text{CaO-}(1-x)\text{TiO}_2)$ as a main component and contains, as a subcomponent, a Li compound in an amount of 2 to 30 parts by weight in terms of Li_2O based on 100 parts by weight of the main component and contains a glass component in an amount of 5 to 150 parts by weight based on 100 parts by weight of the main component.

The calcined body as the main component is obtained by calcining a predetermined amount of the material, which can be represented by the above general formula (2), comprising ZnO, Al_2O_3, SiO_2, and TiO_2, and if necessary, CaO at from 900 to 1200° C. The obtained calcined body contains a $ZnAl_2O_4$ crystal and a Zn_2SiO_4 crystal and further contains a crystal of $CaTiO_3$ when the material composition contains CaO. Crystals of $CaAl_2Si_2O_8$, Zn_2TiO_4, $ZnTiO_3$, $Zn_2Ti_3O_8$, and TiO_2 may be contained, depending on the material composition in some cases.

The glass component to be mixed in the dielectric ceramic composition according to the present embodiment includes a PbO-base glass, a ZnO-base glass, a SiO_2-base glass, a B_2O_3-base glass, as in the case of the first embodiment. Further, as the glass component according to the present embodiment, a glass comprising various metal oxides can be used, and examples thereof include a glass comprising two or more oxides selected from the group consisting of SiO_2, Al_2O_3, ZnO, PbO, Bi_2O_3, BaO, SrO, SnO_2, ZrO_2, and B_2O_3. Either an amorphous glass or a crystalline glass may be used as the glass. When the glass contains PbO, the sintering temperature is liable to lower, however, the unloaded Q-value is liable to decrease and therefore, the content of the PbO component in the glass is preferably 40% by weight or less. A glass containing a ZnO component, an Al_2O_3 component, a BaO component, a SiO_2 component, and a B_2O_3 component is more preferably used as the glass for use in the present embodiment in that a high unloaded Q-value can be obtained.

An example of a glass composition most preferably used includes one containing SiO_2 in an amount of 2.5 to 70 wt %, Al_2O_3 in an amount of 0 to 15 wt %, ZnO in an amount of 10 to 55 wt %, PbO in an amount of 0 to 35 wt %, Bi_2O_3 in an amount of 0 to 2 wt %, BaO in an amount of 0 to 5 wt %, SrO in an amount of 0 to 2 wt %, SnO_2 in an amount of 0 to 2 wt %, ZrO_2 in an amount of 0 to 1 wt %, and B_2O_3 in an amount of 10 to 50 wt %.

Reasons for limiting the composition in the present embodiment will next be described. If the glass component is contained in an amount of less than 5 parts by weight based on 100 parts by weight of the main component served as the base material of the ceramics, a preferable sintered body cannot be obtained at 1000° C. or less; whereas if the glass component is contained in excess of 150 parts by weight, the glass is liable to elute in sintering, with the result that a preferable sintered body cannot be obtained. In the present embodiment, it is most preferable that the glass component be contained in an amount of 10 to 50 parts by weight. This content can lower the sintering temperature and, in particular, makes it harder for a defect such as a migration to occur in the case of simultaneous sintering with Ag which is low melting metal and, at the same time, $Q\times f_0$ value is increased. It is unfavorable that the content of Li_2O to be contained as a subcomponent is less than 2 parts by weight based on 100 parts by weight of the main component, or the content thereof exceeds 30 parts by weight. In the former case, a preferable sintered body cannot be obtained at 1000° C. or less; in the latter case, the glass is liable to elute in sintering with the result that a preferable sintered body cannot be obtained.

It is unfavorable that the molar fraction a in the main component material is less than 7.5 mol %, or it exceeds 55.0 mol %. In the former case, a preferable sintered body cannot be obtained at 1000° C. or less; in the latter case, the $Q\times f_0$ value becomes less than 10000 (GHz). Further, it is unfavorable that the molar fraction b in the main component material is less than 5.0 mol %, or it exceeds 65.0 mol %. In the former case, the $Q\times f_0$ value becomes less than 10000 (GHz); in the latter case, a preferable sintered body cannot be obtained at 1000° C. or less. Further, it is unfavorable that the molar fraction c in the main component material is less than 5.0 mol %, or it exceeds 70.0 mol %. In the former case, a preferable sintered body cannot be obtained at 1000° C. or less; in the latter case, the $Q\times f_0$ value becomes less than 10000 (GHz). Further, it is unfavorable that the molar fraction d in the main component material is less than 7.5 mol % or exceeds 27.5 mol %. In this case, the absolute value in temperature coefficient (τ_f) of resonance frequency becomes more than 20 ppm/° C. Further, it is unfavorable that the value of x exceeds 0.75. In this case, the $Q\times f_0$ value becomes less than 10000 (GHz) and the absolute value in temperature coefficient (τ_f) of resonance frequency becomes more than 20 ppm/° C. The dielectric ceramic composition according to the present embodiment may contain other components in addition, to the main component thereof as far as the object of the present invention is not impaired.

When the value of x is 0 in the above formula (2), the main component material of the dielectric ceramic composition according to the present embodiment is represented by the formula: $a\text{ZnO-}b\text{Al}_2\text{O}_3\text{-}c\text{SiO}_2\text{-}d\text{TiO}_2$ (a+b+c+d=100 mol %). The dielectric ceramic composition containing a Li compound as a subcomponent in an amount of 2 to 30 parts by weight in terms of Li_2O based on 100 parts by weight of the calcined body obtained from the four-phase main component material and a glass component in an amount of 5 to 150 parts by weight based on 100 parts by weight of the main component can also obtain the effect of the present invention.

The dielectric ceramic composition comprising a calcined body obtained from the main component material of the five-phase component represented by ZnO—Al_2O_3—SiO_2—CaO—TiO_2 and Li_2O as a subcomponent in an amount of 2 to 30 parts by weight based on 100 parts by weight of the calcined body and a glass component in an amount of 5 to 150 parts by weight based on 100 parts by weight of the main component can be taken as the most preferable form in the present embodiment.

The dielectric ceramic composition comprising a calcined body obtained from the main component material of the five-phase component, Li_2O as a subcomponent, and a glass component can further lower the sintering temperature and, in particular, makes it harder for a defect such as a migration to occur in the case of simultaneous sintering with Ag which is low melting metal, as compared to the dielectric ceramic composition comprising a calcined body obtained from the main component material of the four-phase component, Li_2O as a subcomponent, and a glass component.

A preferable production method of the dielectric ceramic composition according to the present embodiment and dielectric ceramics obtained by sintering the same will next be described. The respective base materials constituting the main component are obtained as follows. The required base materials of the above ZnO, Al_2O_3, SiO_2, CaO, and TiO_2 are weighed in predetermined amounts and wet-mixed together with a solvent such as water or an alcohol. Subsequently, after removing the water, the alcohol or the like, the obtained powder is calcined at from 900 to 1200° C. Thus-obtained calcined powder of the main component, Li_2O powder as a subcomponent, and a glass powder are weighed in predetermined amounts and wet-mixed together with a solvent such as water or an alcohol. Subsequently, after removing the water, the alcohol or the like, an organic binder such as polyvinyl alcohol and water are mixed in the resulting powder. The mixture is rendered uniform, dried and pulverized, followed by molding under pressure (pressure: on the order of from 100 to 1000 kg/cm^2). The molded product obtained is sintered at from 850 to 975° C. in an oxygen-containing gas atmosphere such as air atmosphere, whereby the dielectric ceramics according to the present embodiment can be obtained.

FIG. **5** shows an X-ray diffraction pattern of the dielectric ceramics according to the present embodiment which is obtained by mixing Li_2O as a subcomponent and glass component with the calcined powder of the main component material of the five-phase component represented by ZnO—Al_2O_3—SiO_2—CaO—TiO_2 and sintering them. As can be seen from FIG. **5**, the dielectric ceramics according to the present embodiment comprises crystalline phases of $ZnAl_2O_4$, Zn_2SiO_4, $CaTiO_3$, $CaAl_2Si_2O_8$ and $Zn_2TiO_3O_8$ and a glass phase.

The dielectric ceramic composition according to the present embodiment is utilized for manufacturing a dielectric resonator, as in the case of the first embodiment. Further, as in the case of the first embodiment, various types of laminated ceramic parts such as a tri-plate type resonator can be obtained from the dielectric ceramic composition according to the present embodiment.

(3) THIRD EMBODIMENT

(Embodiment Related to a Dielectric Ceramic Composition Comprising a Composition Represented by the above General Formula (3) as a Main Component)

A dielectric ceramic composition according to the present embodiment contains a Li compound as a subcomponent in an amount of 2 to 30 parts by weight in terms of Li_2O based on 100 parts by weight of the main component represented by the general formula (3): $aZnAl_2O_4$-bZn_2SiO_4-$cSiO_2$-$dSrTiO_3$ and a glass component in an amount of 5 to 150 parts by weight based on 100 parts by weight of the main component.

The glass component to be mixed in the dielectric ceramic composition according to the present embodiment is the same as that in the second embodiment. When the glass contains PbO, the sintering temperature is liable to lower, however, the unloaded Q-value is liable to decrease and therefore, the content of the PbO component in the glass is preferably 40% by weight or less. A glass containing a ZnO component, an Al_2O_3 component, a BaO component, an SiO_2 component, and a B_2O_3 component is more preferably used as the glass for use in the present embodiment in that a high unloaded Q-value can be obtained.

An example of a glass composition most preferably used includes one containing SiO_2 in an amount of 2.5 to 70 wt %, Al_2O_3 in an amount of 0 to 15 wt %, ZnO in an amount of 10 to 55 wt %, PbO in an amount of 0 to 35 wt %, Bi_2O_3 in an amount of 0 to 2 wt %, BaO in an amount of 0 to 5 wt %, SrO in an amount of 0 to 2 wt %, SnO_2 in an amount of 0 to 2 wt %, ZrO_2 in an amount of 0 to 1 wt %, and B_2O_3 in an amount of 10 to 50 wt %.

Reasons for limiting the composition in the present embodiment will next be described. If the glass component is contained in an amount of less than 5 parts by weight based on 100 parts by weight of the main component served as the base material of the ceramics, a preferable sintered body cannot be obtained at 1000° C. or less; whereas if the glass component is contained in excess of 150 parts by weight, the glass is liable to elute in sintering, with the result that a preferable sintered body cannot be obtained. In the present embodiment, it is more preferable that the glass component be contained in an amount of 20 to 50 parts by weight. This content can lower the sintering temperature and, in particular, makes it harder for a defect such as a migration to occur in the case of simultaneous sintering with Ag which is low melting metal and, at the same time, $Q\times f_0$ value is increased. It is unfavorable that the content of Li compound to be contained as a subcomponent is less than 2 parts by weight in terms of Li_2O, based on 100 parts by weight of the main component, or it exceeds 30 parts by weight. In the former case, a preferable sintered body cannot be obtained at 1000° C. or less; in the latter case, the glass is liable to elute in sintering with the result that a preferable sintered body cannot be obtained.

It is unfavorable that the molar fraction a in the main component is less than 2.5 mol %, or it exceeds 77.5 mol %. In the former case, the $Q\times f_0$ value becomes less than 10000 (GHz); in the latter case, a preferable sintered body cannot be obtained at 1000° C. or less. Further, it is unfavorable that the molar fraction b in the main component is less than 2.5 mol %, or it exceeds 77.5 mol %. In the former case, a preferable sintered body cannot be obtained at 1000° C. or less; in the latter case, the $Q\times f_0$ value becomes less than 10000 (GHz). Further, it is unfavorable that the molar fraction c in the main component is less than 2.5 mol %, or it exceeds 37.5 mol %. In the former case, a preferable sintered body cannot be obtained at 1000° C. or less; in the latter case, the $Q\times f_0$ value becomes less than 10000 (GHz). Further, it is unfavorable that the molar fraction d in the main component is less than 10.0 mol % or exceeds 17.5 mol %. In this case, the absolute value in temperature coefficient (τ_f) of resonance frequency becomes more than 20 ppm/° C. The dielectric ceramic composition may contain other components in addition to the main component thereof as far as the object of the present invention is not impaired.

A preferable production method of the dielectric ceramic composition according to the present embodiment and dielectric ceramics obtained by sintering the same will next be described. The respective components of $ZnAl_2O_4$, Zn_2SiO_4, SiO_2, $SrTiO_3$ that constitute the main component of the dielectric ceramic composition according to the present embodiment can be prepared individually, or can be prepared at a time as a mixture. When the respective components are individually prepared, oxides of each element are mixed in a predetermined ratio, followed by calcination. For example, $ZnAl_2O_4$ is obtained by mixing ZnO and Al_2O_3 in a molar ratio of 1:1 followed by calcination at from 900 to 1200° C. Similarly, Zn_2SiO_4 is obtained by mixing ZnO and SiO_2 in a molar ratio of 2:1 followed by calcination. $SrTiO_3$ is obtained by mixing SrO and TiO_2 in a molar ratio of 1:1 followed by calcination.

When the main component of the dielectric ceramic composition is to be prepared at a time as a mixture, it can be obtained as follows. That is, predetermined amounts of the required starting materials of respective powders: zinc oxide (ZnO), aluminium oxide (Al_2O_3), silicon oxide (SiO_2), strontium oxide (SrO), titanium oxide (TiO_2) are wet-mixed together with a solvent such as water or an alcohol. Subsequently, after removing the water, the alcohol, or the like, the mixture is calcined at from 900 to 1200° C. for 2 hours in air atmosphere to obtain a calcined powder comprising $ZnAl_2O_4$, Zn_2SiO_4, SiO_2, and $SrTiO_3$. As materials of zinc, aluminium, silicon, strontium, and titanium, carbonate, hydroxide, and an organic metal compound each turning into an oxide at the time of calcining, may also be used, in addition to ZnO, Al_2O_3, SiO_2, SrO, and TiO_2.

The Li_2O powder as a subcomponent and glass powder are mixed with the main component comprising the calcined powder obtained in the manner as described above, whereby obtaining the dielectric ceramic composition according to the present embodiment. In the present embodiment, the same amount in terms of Li_2O of a compound turning into Li_2O at the time of calcining, such as Li_2CO_3 can be used in place of Li_2O.

When the dielectric ceramics is to be obtained by sintering the dielectric ceramic composition, the following procedure is required. That is, $ZnAl_2O_4$, Zn_2SiO_4, SiO_2, and $SrTiO_3$ as a main component, a Li_2O powder as a subcomponent and a glass powder are weighed in predetermined amounts and wet-mixed together with a solvent such as water or an alcohol. Subsequently, after removing the water, the alcohol or the like, an organic binder such as polyvinyl alcohol and water are mixed in the resulting powder. The mixture is rendered uniform, dried and pulverized, followed by molding under pressure (pressure: on the order of from 100 to 1000 kg/cm^2). The molded product obtained is sintered at from 825 to 975° C. in an oxygen-containing gas atmosphere such as air atmosphere, whereby the dielectric ceramics comprising crystalline phases of $ZnAl_2O_4$, Zn_2SiO_4, and $SrTiO_3$ and a glass phase can be obtained.

FIG. **6** shows an X-ray diffraction pattern of the dielectric ceramics according to the present embodiment which is obtained by mixing Li_2O as a subcomponent and glass component with the calcined powder of the main component represented by $aZnAl_2O_4$-bZn_2SiO_4-$cSiO_2$-$dSrTiO_3$ and sintering them. As can be seen from FIG. **6**, the dielectric ceramics according to the present embodiment comprises crystalline phases of $ZnAl_2O_4$, Zn_2SiO_4, and $SrTiO_3$ and a glass phase. A SiO_2 crystal in the main component performs an important role in sintering. When the SiO_2 crystal is not contained in the main component, the mixture cannot be sintered sufficiently at a low temperature. It is estimated that the SiO_2 crystal contained in the main component before sintering, which is not detected by the X-ray diffraction after sintering, becomes amorphous after sintering. Similarly, Li_2O as a subcomponent is not detected by the X-ray diffraction after sintering and is estimated to become amorphous after sintering. The dielectric ceramics according to the present embodiment comprises the crystalline phases of $ZnAl_2O_4$, Zn_2SiO_4, and $SrTiO_3$, tiny amount of another crystalline phase may exist as far as the effects of the present invention are not impaired. It is possible to obtain the above crystalline structure by the production method in which the oxides are used as starting materials, as well as by using the production method in which carbonate, hydroxide, and an organic metal compound each turning into an oxide at the time of calcining are used as the stating materials.

The dielectric ceramic composition according to the present embodiment is utilized for manufacturing a dielectric resonator, as in the case of the first embodiment. Further, as in the case of the first embodiment, various types of laminated ceramic parts such as a tri-plate type resonator can be obtained from the dielectric ceramic composition according to the present embodiment.

(4) FOURTH EMBODIMENT

(Embodiment Related to a Dielectric Ceramic Composition Comprising a Composition Represented by the above General Formula (4) as a Main Component)

A dielectric ceramic composition according to the present embodiment contains a Li compound as a subcomponent in an amount of 1 to 15 parts by weight in terms of Li_2O based on 100 parts by weight of the main component represented by the general formula (4): aMg_2SiO_4-$bZnAl_2O_4$-$cSiO_2$-$dCaTiO_3$-eZn_2SiO_4 and a glass component in an amount of 5 to 150 parts by weight based on 100 parts by weight of the main component.

It is unfavorable that the molar fraction a in the main component is less than 0.10, or it exceeds 0.72. In the former case, a preferable sintered body cannot be obtained at 1000° C. or less; in the latter case, the $Q\times f_0$ value becomes less than 10000 (GHz). Further, it is unfavorable that the molar fraction b in the main component is less than 0.08, or it exceeds 0.62. In the former case, the $Q\times f_0$ value becomes less than 10000 (GHz); in the latter case, a preferable sintered body cannot be obtained at 1000° C. or less. Further, it is unfavorable that the molar fraction c in the main component is less than 0.02, or it exceeds 0.22. In the former case, a preferable sintered body cannot be obtained at 1000° C. or less; in the latter case, the $Q\times f_0$ value becomes less than 10000 (GHz). Further, it is unfavorable that the molar fraction d in the main component is less than 0.12 or exceeds 0.22. In this case, the absolute value in temperature coefficient (of) of resonance frequency becomes more than 20 ppm/° C. Further, it is unfavorable that the molar fraction e in the main component exceeds 0.08. In this case, the $Q\times f_0$ value becomes less than 10000 (GHz). The dielectric ceramic composition according to the present embodiment may contain other components in addition to the main component thereof as far as the object of the present invention is not impaired.

In the dielectric ceramic composition according to the present embodiment, the Li compound used as a subcomponent includes Li_2O, as well as carbonate, hydroxide, and an organic metal compound each turning into an oxide at the time of calcining. In general, Li_2O or Li_2CO_3 is used. The amount to be added of the subcomponent is determined as above in terms of Li_2O.

As in the case of the first embodiment, the glass component to be mixed in the dielectric ceramic composition according to the present embodiment includes a PbO-base glass, a ZnO-base glass, a SiO_2-base glass, a B_2O_3-base glass. In addition to the above, a glass comprising various metal oxides can also be used, and examples thereof include a glass comprising two or more oxides selected from the group consisting of SiO_2, Al_2O_3, ZnO, PbO, Bi_2O_3, BaO, SrO, SnO_2, ZrO_2, and B_2O_3. Either an amorphous glass or a crystalline glass may be used as the glass. When the glass contains PbO, the sintering temperature is liable to lower, however, the unloaded Q-value is liable to decrease and therefore, the content of the PbO component in the glass is preferably 40% by weight or less. A glass containing a ZnO component, an Al_2O_3 component, a BaO component, an SiO_2 component, and a B_2O_3 component is more preferably used as the glass for use in the present embodiment in that a high unloaded Q-value can be obtained.

An example of a glass composition most preferably used includes one containing SiO_2 in an amount of 2 to 70 wt %, Al_2O_3 in an amount of 0 to 15 wt %, ZnO in an amount of 10 to 55 wt %, PbO in an amount of 0 to 35 wt %, Bi_2O_3 in an amount of 0 to 2 wt %, BaO in an amount of 0 to 30 wt %, SrO in an amount of 0 to 2 wt %, SnO_2 in an amount of 0 to 2 wt %, ZrO_2 in an amount of 0 to 1 wt %, and B_2O_3 in an amount of 10 to 50 wt %. The use of the above glass composition allows the sintering at a low temperature of 950° C. or less.

If the glass component is contained in an amount of less than 5 parts by weight based on 100 parts by weight of the main component served as the base material of the ceramics, a preferable sintered body cannot be obtained at 1000° C. or less; whereas if the glass component is contained in excess of 150 parts by weight, the glass is liable to elute in sintering, with the result that a preferable sintered body cannot be obtained. In the present embodiment, it is most preferable that the glass component be contained in an amount of 10 to 50 parts by weight. This content can lower the sintering temperature and, in particular, makes it harder for a defect such as a migration to occur in the case of simultaneous sintering with Ag which is low melting metal and, at the same time, $Q\times f_0$ value is increased. It is unfavorable that the content of the subcomponent is less than 1 part by weight in terms of Li_2O based on 100 parts by weight of the main component, or it exceeds 15 parts by weight. In the former case, a preferable sintered body cannot be obtained at 1000° C. or less; in the latter case, the glass is liable to elute in sintering with the result that a preferable sintered body cannot be obtained.

When the molar fraction e of the main component is 0, the main component of the dielectric ceramic composition according to the present embodiment is represented by the formula: aMg_2SiO_4-$bZnAl_2O_4$-$cSiO_2$-$dCaTiO_3$, and the respective molar fractions of a, b, c, and d are represented by: $0.10\leqq a\leqq 0.72$, $0.08\leqq b\leqq 0.62$, $0.02\leqq c\leqq 0.22$, and $0.12\leqq d\leqq 0.22$ ($a+b+c+d=1$). The dielectric ceramic composition comprising a Li compound to be contained as a subcomponent in an amount of 1 to 15 parts by weight in terms of Li_2O and a glass component in an amount of 5 to 150 parts by weight, based on 100 parts by weight of the four-phase main component can also obtain the effect of the present invention.

The dielectric ceramic composition comprising a five-phase main component: aMg_2SiO_4-$bZnAl_2O_4$-$cSiO_2$-$dCaTiO_3$-eZn_2SiO_4 and a Li compound to be contained as a subcomponent in an amount of 1 to 15 parts by weight in terms of Li_2O based on 100 parts by weight of the main component and glass component in an amount of 5 to 150 parts by weight based on 100 parts by weight of the main component can be taken as the most preferable form in the present embodiment. The addition of the Zn_2SiO_4 to the dielectric ceramic composition comprising the four-phase main component can lower the sintering temperature and, in particular, makes it harder for a defect such as a migration to occur in the case of simultaneous sintering with Ag which is low melting metal.

The dielectric ceramics according to the present embodiment is obtained by sintering the dielectric ceramic composition. The obtained dielectric ceramics comprises crystalline phases of Mg_2SiO_4, $ZnAl_2O_4$, SiO_2, and $CaTiO_3$, and a glass phase. When the dielectric ceramic composition before sintering comprises Zn_2SiO_4, the obtained dielectric ceramics further comprises Zn_2SiO_4 as a crystalline phase. Although the compositions of the crystalline phase and glass phase substantially correspond to those constituting the dielectric ceramic composition, the surface of crystal grains and glass component are partly reacted to form a strong sintered body, as well as, the crystal component and glass component are partly reacted to generate at least one of the crystals of Zn_2SiO_4, Li_2ZnSiO_4, $CaTiSiO_5$, Ca_2TiSiO_6, $BaAl_2Si_2O_8$, and $Zn_2Ti_3O_8$, in some cases.

The dielectric ceramics according to the present embodiment exhibits a large $Q\times f_0$ value of 20000 (GHz) or more, which is a product of resonance frequency f_0 (GHz) and Q-value, and has a low dielectric loss. Further, the absolute value in temperature coefficient (τf) of resonance frequency is not more than 20 ppm/° C., so that the influence of the temperature can be reduced. Further, dielectric constant ϵ_r is not more than 10, so that a high-frequency device or high-frequency circuit obtained using the dielectric ceramics is not excessively reduced in the size, but can be kept in an appropriate size. As a result, the dielectric ceramics according to the present embodiment is excellent in processing accuracy and productivity.

A preferable production method of the dielectric ceramic composition according to the present embodiment and dielectric ceramics obtained by sintering the same will next be described. The respective components of Mg_2SiO_4, $ZnAl_2O_4$, SiO_2, $CaTiO_3$, and Zn_2SiO_4 that constitute the main component of the dielectric ceramic composition according to the present embodiment can be prepared individually, or can be prepared at a time as a mixture. When the respective components are individually prepared, oxides of each element are mixed in a predetermined ratio, followed by calcination. For example, Mg_2SiO_4 is obtained by mixing MgO and SiO_2 in a molar ratio of 2:1 followed by calcination at from 900 to 1300° C. $ZnAl_2O_4$ is obtained by mixing ZnO and Al_2O_3 in a molar ratio of 1:1 followed by calcination at from 900 to 1300° C. $CaTiO_3$ is obtained by mixing CaO and TiO_2 in a molar ratio of 1:1 followed by calcination. Zn_2SiO_4 is obtained by mixing ZnO and SiO_2 in a molar ratio of 2:1 followed by calcination.

When the main component of the dielectric ceramic composition is to be prepared at a time as a mixture, it can be obtained as follows. That is, predetermined amounts of the required starting materials of respective powders: magnesium oxide (MgO), zinc oxide (ZnO), aluminium oxide (Al_2O_3), silicon oxide (SiO_2), calcium oxide (CaO), and titanium oxide (TiO_2) are wet-mixed together with a solvent such as water or an alcohol. Subsequently, after removing the water the alcohol, or the like, the mixture is calcined at from 900 to 1300° C. for 2 hours in air atmosphere to obtain a calcined powder comprising Mg_2SiO_4, $ZnAl_2O_4$, SiO_2, $CaTiO_3$, and Zn_2SiO_4. As materials of magnesium, zinc, aluminium, silicon, calcium, and titanium, carbonate, hydroxide, and an organic metal compound each turning into an oxide at the time of calcining, may also be used, in addition to MgO, ZnO, Al_2O_3, SiO_2, CaO, and TiO_2.

A Li compound turning into Li_2O at the time of calcining such as lithium carbonate (Li_2CO_3) powder as a subcomponent and glass powder are mixed with the main component comprising the calcined powder obtained in the manner as described above, whereby the dielectric ceramic composition according to the present embodiment can be obtained.

When the dielectric ceramics is to be obtained by sintering the dielectric ceramic composition, the following procedure is required. That is, the required main component powders of Mg_2SiO_4, $ZnAl_2O_4$, SiO_2, $CaTiO_3$, and Zn_2SiO_4 and a powder of a Li compound, as a subcomponent, turning into Li_2O at the time of calcining and glass powder are weighed in predetermined amounts and wet-mixed together with a solvent such as water or an alcohol. Subsequently, after removing the water, the alcohol or the like, an organic binder such as polyvinyl alcohol and water are mixed in the resulting powder. The mixture is rendered uniform, dried and pulverized, followed by molding under pressure (pressure: on the order of from 100 to 1000 kg/cm^2). The molded product obtained is sintered at from 800 to 950° C. in an oxygen-containing gas atmosphere such as air atmosphere, whereby the dielectric ceramics comprising crystalline phases of Mg_2SiO_4, $ZnAl_2O_4$, SiO_2, $CaTiO_3$, and a glass phase can be obtained. In some cases, the dielectric ceramics comprises at least one of the crystalline phases of Li_2ZnSiO_4, $CaTiSiO_5$, Ca_2TiSiO_6, $BaAl_2Si_2O_8$, and $Zn_2Ti_3O_8$, due to a reaction between the main component and subcomponent, or between the main component and glass component. These crystalline phases are optional components and, even if one of these exists, the effect of the present invention is not impaired.

FIG. **7** shows an X-ray diffraction pattern of the dielectric ceramics according to the present embodiment which is obtained by mixing Li_2O as a subcomponent and glass component with the above calcined powder of the four-phase main component represented by aMg_2SiO_4-$bZnAl_2O_4$-$cSiO_2$-$dCaTiO_3$ and sintering them. FIG. **8** shows an X-ray diffraction pattern of the dielectric ceramics according to the present embodiment which is obtained by mixing the Li_2O as a subcomponent and glass component with the calcined powder of the five-phase main component represented by aMg_2SiO_4-$bZnAl_2O_4$-$cSiO_2$-$dCaTiO_3$-eZn_2SiO_4 and sintering them.

As shown in FIGS. **7** and **8**, the dielectric ceramics according to the present embodiment comprise at least one of the crystalline phases of Li_2ZnSiO_4, $CaTiSiO_5$, Ca_2TiSiO_6, $BaAl_2Si_2O_8$, and $Zn_2Ti_3O_8$, due to a reaction between the main component and subcomponent, or between the main component and glass component in addition to the crystalline phases of the main component containing Mg_2SiO_4, $ZnAl_2O_4$, SiO_2, and $CaTiO_3$, (in the case of FIG. **8**, further containing Zn_2SiO_4) and glass phase. However, even if such a crystalline phase exists in the dielectric ceramic composition according to the present embodiment, the effect of the present invention is not impaired. Tiny amount of another crystalline phase may exist in addition to the above crystalline phases, as far as the effects of the present invention are not impaired. It is possible to obtain the above crystalline structure by the production method in which the oxides of each element are used as starting materials, as well as by using the production method in which carbonate, hydroxide, and an organic metal compound each turning into an oxide at the time of calcining are used as the stating materials.

The dielectric ceramic composition according to the present embodiment is utilized for manufacturing a dielectric resonator, as in the case of the first embodiment. Further, as in the case of the first embodiment, various types of laminated ceramic parts such as a tri-plate type resonator can be obtained from the dielectric ceramic composition according to the present embodiment.

Hereinafter, examples according to the present invention will be described.

(1) EXAMPLES AND COMPARATIVE EXAMPLES RELATED TO THE FIRST EMBODIMENT

Example 1

Respective powders of ZnO and Al_2O_3 were weighed so that the molar ratio between them became 1:1. The weighed powders were charged into a ball mill together with ethanol (solvent, the same in the following description) and ZrO_2 ball and wet-mixed for 24 hours. After removing the solvent from the solution, the resulting mixture was dried and calcined in air atmosphere at 1000° C. for 2 hours to obtain a powder of $ZnAl_2O_4$ crystal.

Similarly, respective powders of ZnO and SiO_2 were weighed so that the molar ratio between them became 2:1. The weighed powders were charged into a ball mill together with ethanol and ZrO_2 ball and wet-mixed for 24 hours. After removing the solvent from the solution, the resulting mixture was dried and calcined in air atmosphere at 1000° C. for 2 hours to obtain a powder of Zn_2SiO_4 crystal.

Subsequently, the powders of $ZnAl_2O_4$ and Zn_2SiO_4 thus obtained and TiO_2 powder were weighed in amounts of 7.5 mol %, 67.5 mol %, and 25 mol %, respectively. The weighed powders were then mixed to obtain the main component of the dielectric ceramic composition.

Powders of the main component and a glass powder were weighed in predetermined amounts (total amount of 150 g) so that the glass powder containing SiO_2 in an amount of 6.0 wt %, Al_2O_3 in an amount of 11.0 wt %, ZnO in an amount of 47.0 wt %, BaO in an amount of 4.0 wt %, SrO in an amount 0.2 wt %, CaO in an amount of 0.8 wt %, SnO_2 in an amount of 1.0 wt %, and B_2O_3 in an amount of 30.0 wt % became 30 parts by weight based on 100 parts by weight of the main component. The weighed powders were charged into a ball mill together with ethanol and ZrO_2 ball and wet-mixed for 24 hours. After removing the solvent from the solution, the resulting mixture was dried.

The obtained mixed powder (composition thereof is shown in Table 1) was pulverized. Then, to the pulverized product, an appropriate amount of a polyvinyl alcohol solution was added, followed by drying. Thereafter, the resulting pulverized product was molded into a pellet having a diameter of 10 mm and a thickness of 5 mm and the pellet obtained was sintered in air atmosphere at 925° C. for 2 hours, whereby the dielectric ceramics having the composition according to the present invention was obtained.

The thus-obtained dielectric ceramics were processed to a size of 8 mm in diameter and 4 mm in thickness and then measured by a dielectric resonance method to calculate the $Q \times f_0$ value at the resonance frequency of 9 to 13 GHz, the dielectric constant ϵ_r and the temperature coefficient τ_f of resonance frequency. The results thereof are shown in Table 2.

Also, to 100 g of the dry-mixed powder obtained by mixing the main component and the glass powder followed by solvent removal, 9 g of polyvinyl butyral as a binder, 6 g of dibutylphthalate as a plasticizer, and 60 g of toluene and 30 g of isopropyl alcohol both as a solvent were added to produce a green sheet having a thickness of 100 μm by the doctor blade method. Then, 20 layers of the green sheets were laminated by the thermo compression bonding of applying a pressure of 200 kg/cm^2 at a temperature of 65° C. At this time, a layer having been printed with Ag pattern as an internal electrode was disposed such that it was provided at the center in the thickness direction. After sintering the obtained laminated product at 925° C. for 2 hours, the sintered body was processed to a size of 5.0 mm in width, 1.5 mm in height (dimension in laminated direction) and 9.5 mm in length (dimension in extending direction of internal electrode), and an external electrode was formed to produce a tri-plate type resonator, as shown in FIGS. **1** and **2** The obtained tri-plate type resonator was evaluated on the unloaded Q-value at a resonance frequency of 2.5 GHz. The result thereof is shown in Table 2.

Examples 2 to 10

In the same manner as in Example 1, $ZnAl_2O_4$ and Zn_2SiO_4 powders, TiO_2 powder, and glass powder were weighed to have the composition ratios shown in Table 1, respectively. The weighed powders were then mixed and molded under the same condition as Example 1. The pellets obtained were sintered in air atmosphere at from 900 to 925° C. for 2 hours as shown in Table 2, whereby dielectric ceramics and resonators were obtained. The obtained dielectric ceramics and resonators were evaluated on various properties in the same method as in Example 1. The results thereof are shown in Table 2. Note that FIG. **3** shows the X-ray diffraction pattern of the dielectric ceramics obtained in Example 2.

Comparative Examples 1 to 8

In the same manner as in Example 1, $ZnAl_2O_4$ and Zn_2SiO_4 powders, TiO_2 powder, and glass powder were weighed to have the composition ratios shown in Table 1, respectively. The weighed powders were then mixed and molded under the same condition as Example 1. The pellets obtained were sintered in air atmosphere at from 905 to 1000° C. for 2 hours as shown in Table 2, whereby dielectric ceramics and resonators were obtained (preferable sintered bodies were not obtained in some comparative examples). The obtained dielectric ceramics and resonators were evaluated on various properties in the same method as in Example 1. The results thereof are shown in Table 2.

Example 11

Respective powders of ZnO and TiO_2 were weighed so that the molar ratio between them became 2:1. The weighed powders were charged into a ball mill together with ethanol and ZrO_2 ball and wet-mixed for 24 hours. After removing the solvent from the solution, the resulting mixture was dried and calcined in air atmosphere at 1000° C. for 2 hours to obtain a powder of Zn_2TiO_4 crystal.

Further, in the same manner as in Example 1, $ZnAl_2O_4$ and Zn_2SiO_4 powders were obtained.

The $ZnAl_2O_4$ powder, Zn_2SiO_4 powder, and Zn_2TiO_4 powder thus obtained and TiO_2 powder and glass powder were weighed to have the composition ratio shown in Table 1, respectively. The weighed powders were then mixed and molded under the same condition as Example 1. The pellet obtained was sintered in air atmosphere at 875° C. for 2 hours, whereby dielectric ceramics and resonators were obtained. The obtained dielectric ceramics and resonators were evaluated on various properties in the same method as in Example 1. The results thereof are shown in Table 2.

Examples 12 to 22

In the same manner as in Example 11, $ZnAl_2O_4$, Zn_2SiO_4, and Zn_2TiO_4 powders, TiO_2 powder, and glass powder were weighed to have the composition ratios shown in Table 1, respectively. The weighed powders were then mixed and molded under the same condition as Example 1. The pellets obtained were sintered in air atmosphere at from 825 to 905° C. for 2 hours as shown in Table 2, whereby dielectric ceramics and resonators were obtained. The obtained dielectric ceramics and resonators were evaluated on various properties in the same method as in Example 1. The results thereof are shown in Table 2. Note that FIG. **4** shows the X-ray diffraction pattern of the dielectric ceramics obtained in Example 13.

Comparative Examples 9 to 18

In the same manner as in Example 11, $ZnAl_2O_4$, Zn_2SiO_4, and Zn_2TiO_4 powders, TiO_2 powder, and glass powder were weighed to have the composition ratios shown in Table 1, respectively. The weighed powders were then mixed and molded under the same condition as Example 1. The pellets obtained were sintered in air atmosphere at from 850 to 1000° C. for 2 hours as shown in Table 2, whereby dielectric ceramics and resonators were obtained (preferable sintered bodies were not obtained in some comparative examples). The obtained dielectric ceramics and resonators were evaluated on various properties in the same method as in Example 1. The results thereof are shown in Table 2.

(2) EXAMPLES AND COMPARATIVE EXAMPLES RELATED TO THE SECOND EMBODIMENT

Example 23

Predetermined amounts (200 g, in total) of ZnO, Al_2O_3, SiO_2, and TiO_2 were weighed in an amount of 10.0 mol %, 35.0 mol %, 35.0 mol %, and 20.0 mol %, respectively. The weighed material powders were charged into a ball mill together with ethanol and ZrO_2 ball and wet-mixed for 24 hours. After removing the solvent from the solution, the resulting mixed powder was pulverized. Then, the pulverized product was calcined at 1000° C. in air atmosphere. The obtained calcined powder was pulverized to obtain the main component.

Powders of the main component and a glass powder were weighed in predetermined amounts (150 g, in total) so that Li_2O powder and the glass powder containing SiO_2 in an amount of 6.0 wt %, Al_2O_3 in an amount of 12.0 wt %, ZnO in an amount of 47.0 wt %, BaO in an amount of 3.0 wt %, SrO in an amount of 1.0 wt %, SnO_2 in an amount of 1.0 wt %, and B_2O_3 in an amount of 30.0 wt % became 5 parts by weight and 20 parts by weight, respectively, based on 100 parts by weight of the main component. The weighed powders were charged into a ball mill together with ethanol and ZrO_2 ball and wet-mixed for 24 hours. After removing the solvent from the solution, the resulting mixture was dried.

The obtained mixed powder was pulverized. Then, to the pulverized product, an appropriate amount of a polyvinyl alcohol solution was added, followed by drying. Thereafter, the resulting pulverized product was molded into a pellet having a diameter of 10 mm and a thickness of 5 mm and the pellet obtained was sintered in air atmosphere at 925° C. for 2 hours, whereby the dielectric ceramics was obtained.

The thus-obtained dielectric ceramics was processed to a size of 8 mm in diameter and 4 mm in thickness and then measured by a dielectric resonance method to calculate the $Q \times f_0$ value at the resonance frequency of 9 to 13 GHz, the dielectric constant ϵ_r and the temperature coefficient τ_f of resonance frequency. The results thereof are shown in Table 4.

Also, to 100 g of the dry-mixed powder obtained by mixing the main component, Li_2O powder, and the glass powder followed by solvent removal, 9 g of polyvinyl butyral as a binder, 6 g of dibutylphthalate as a plasticizer, and 60 g of toluene and 30 g of isopropyl alcohol both as a solvent were added to produce a green sheet having a thickness of 100 μm by the doctor blade method. Then, 20 layers of the green sheets were laminated by the thermo compression bonding of applying a pressure of 200 kg/cm^2 at a temperature of 65° C. At this time, a layer having been printed with Ag pattern as an internal electrode was disposed such that it was provided at the center in the thickness direction. After sintering the obtained laminated product at 975° C. for 2 hours, the sintered body was processed to a size of 5.0 mm in width, 1.5 mm in height and 9.5 mm in length, and an external electrode was formed to produce a tri-plate type resonator, as shown in FIGS. **1** and **2**. The obtained tri-plate type resonator was evaluated on the unloaded Q-value at a resonance frequency of 2.5 GHz. The result thereof is shown in Table 4.

Examples 24 to 42

In the same manner as in Example 23, the calcined powder of the main component, Li_2O powder, and glass powder were weighed to have the composition ratios shown in Table 3, respectively. The weighed powders were then mixed and molded under the same condition as Example 23. The pellets obtained were sintered in air atmosphere at from 850 to 975° C. for 2 hours as shown in Table 3, whereby dielectric ceramics and resonators were obtained. The obtained dielectric ceramics and resonators were evaluated on various properties in the same method as in Example 23. The results thereof are shown in Table 4. Note that FIG. **5** shows the X-ray diffraction pattern of the dielectric ceramics obtained in Example 33.

Comparative Examples 19 to 29

In the same manner as in Example 23, the calcined powder of the main component, Li_2O powder, and glass powder were weighed to have the composition ratios shown in Table 3, respectively. The weighed powders were then mixed and molded under the same condition as Example 23. The pellets obtained were sintered in air atmosphere at from 875 to 1000° C. for 2 hours as shown in Table 4, whereby dielectric ceramics and resonators were obtained (preferable sintered bodies were not obtained in some comparative examples). The obtained dielectric ceramics and resonators were evaluated on various properties in the same method as in Example 23. The results thereof are shown in Table 4.

(3) EXAMPLES AND COMPARATIVE EXAMPLES RELATED TO THE THIRD EMBODIMENT

Example 43

In order to obtain a main component containing $ZnAl_2O_4$ in an amount of 75.0 mol %, Zn_2SiO_4 in an amount of 5.0 mol %, SiO_2 in an amount of 5.0 mol %, and $SrTiO_3$ in an amount of 15.0 mol %, predetermined amounts (200 g, in total) of the respective powders including zinc oxide (ZnO), aluminium oxide (Al_2O_3), silicon oxide (SiO_2), strontium oxide (SrO), and titanium oxide (TiO_2) were weighed. The weighed powders were charged into a ball mill together with ethanol and ZrO_2 ball and wet-mixed for 24 hours. After removing the solvent from the solution, the resulting mixed powder was pulverized. Then, the pulverized product was calcined at 1000° C. in air atmosphere. The obtained calcined powder was pulverized to obtain the main component.

Powder of the main component, Li_2O powder and a glass powder were weighed in predetermined amounts (150 g, in total) so that Li_2O powder and the glass powder containing SiO_2 in an amount of 6.0 wt %, Al_2O_3 in an amount of 12.0 wt %, ZnO in an amount of 47.0 wt %, BaO in an amount of 3.0 wt %, SrO in an amount of 1.0 wt %, SnO_2 in an amount of 1.0 wt %, and B_2O_3 in an amount of 30.0 wt % became 5 parts by weight and 25 parts by weight, respectively, based on 100 parts by weight of the main component. The weighed powders were charged into a ball mill together with ethanol and ZrO_2 ball and wet-mixed for 24 hours. After removing the solvent from the solution, the resulting mixture was dried.

The obtained mixed powder was pulverized. Then, to the pulverized product, an appropriate amount of a polyvinyl alcohol solution was added, followed by drying. Thereafter, the resulting pulverized product was molded into a pellet having a diameter of 10 mm and a thickness of 5 mm and the pellet obtained was sintered in air atmosphere at 975° C. for 2 hours, whereby the dielectric ceramics was obtained.

The thus-obtained dielectric ceramics was processed to a size of 8 mm in diameter and 4 mm in thickness and then measured by a dielectric resonance method to calculate the $Q \times f_0$ value at the resonance frequency of 9 to 13 GHz, the dielectric constant ϵ_r and the temperature coefficient τ_f of resonance frequency. The results thereof are shown in Table 6.

Also, to 100 g of the dry-mixed powder obtained by mixing the main component, Li_2O powder, and the glass powder followed by solvent removal, 9 g of polyvinyl butyral as a binder, 6 g of dibutylphthalate as a plasticizer, and 60 g of toluene and 30 g of isopropyl alcohol both as a solvent were added to produce a green sheet having a thickness of 100 μm by the doctor blade method. Then, 20 layers of the green sheets were laminated by the thermo compression bonding of applying a pressure of 200 kg/cm^2 at a temperature of 65° C. At this time, a layer having been printed with Ag pattern as an internal electrode was disposed such that it was provided at the center in the thickness direction. After sintering the obtained laminated product at 975° C. for 2 hours, the sintered body was processed to a size of 5.0 mm in width, 1.5 mm in height and 9.5 mm in length, and an external electrode was formed to produce a tri-plate type resonator, as shown in FIGS. **1** and **2**. The obtained tri-plate type resonator was evaluated on the unloaded Q-value at a resonance frequency of 2.5 GHz. The result thereof is shown in Table 6.

Examples 44 to 62

In the same manner as in Example 43, the calcined powder of the main component, Li_2O powder, and glass powder were weighed to have the composition ratios shown in Table 5, respectively. The weighed powders were then mixed and molded under the same condition as Example 43. The pellets obtained were sintered in air atmosphere at from 825 to 975° C. for 2 hours as shown in Table 6, whereby dielectric ceramics and resonators were obtained. The obtained dielectric ceramics and resonators were evaluated on various properties in the same method as in Example 43. The results thereof are shown in Table 6. FIG. **6** shows the X-ray diffraction pattern of the dielectric ceramics of Example 44, which is obtained by sintering the dielectric ceramic composition obtained by mixing the main component represented by $aZnAl_2O_4$-bZn_2SiO_4-$cSiO_2$-$dSrTiO_3$ according to the present invention, Li_2O as a subcomponent, and glass component.

Comparative Examples 30 to 41

In the same manner as in Example 43, the calcined powder of the main component, Li_2O powder, and glass powder were weighed to have the composition ratios shown in Table 5, respectively. The weighed powders were then mixed and molded under the same condition as Example 43. The pellets obtained were sintered in air atmosphere at from 825 to 1000° C. for 2 hours as shown in Table 6, whereby dielectric ceramics and resonators were obtained (preferable sintered bodies were not obtained in some comparative examples). The obtained dielectric ceramics and resonators were evaluated on various properties in the same method as in Example 43. The results thereof are shown in Table 6.

(4) EXAMPLES AND COMPARATIVE EXAMPLES RELATED TO THE FOURTH EMBODIMENT

Example 63

MgO and SiO_2 were weighed so that the molar ratio between them became 2:1. The weighed powders were charged into a ball mill together with ethanol and ZrO_2 ball and wet-mixed for 24 hours. After removing the solvent from the solution, the resulting mixture was dried and calcined at 1200° C. for 2 hours. The obtained calcined body was pulverized to obtain a powder of Mg_2SiO_4. Further, ZnO and Al_2O_3 were weighed so that the molar ratio between them became 1:1. The weighed powders were charged into a ball mill together with ethanol and ZrO_2 ball and wet-mixed for 24 hours. After removing the solvent from the solution, the resulting mixture was dried and calcined at 1100° C. for 2 hours. The obtained calcined body was pulverized to obtain a powder of $ZnAl_2O_4$. Further, CaO and TiO_2 were weighed so that the molar ratio between them became 1:1. The weighed powders were charged into a ball mill together with ethanol and ZrO_2 ball and wet-mixed for 24 hours. After removing the solvent from the solution, the resulting mixture was dried and calcined at 1250° C. for 2 hours. The obtained calcined body was pulverized to obtain a powder of $CaTiO_3$.

Then, Mg_2SiO_4, $ZnAl_2O_4$, SiO_2, and $CaTiO_3$ were weighed in molar fractions of 0.12, 0.58, 0.10, and 0.20, respectively and mixed to obtain the main component. Further, 100 parts by weight of the main component, 5 parts by weight of Li_2O powder, and 25 parts by weight of glass powder containing SiO_2 in an amount of 6.0 wt %, Al_2O_3 in an amount of 12.0 wt %, ZnO in an amount of 25.0 wt %, BaO in an amount of 25.0 wt %, SrO in an amount of 1.0 wt %, SnO_2 in an amount of 1.0 wt %, and B_2O_3 in an amount of 30.0 wt % were weighed in predetermined amounts (150 g, in total). The weighed powders were charged into a ball mill together with ethanol and ZrO_2 ball and wet-mixed for 24 hours. After removing the solvent from the solution, the resulting mixture was dried.

The obtained mixed powder was pulverized. Then, to the pulverized product, an appropriate amount of a polyvinyl alcohol solution was added, followed by drying. Thereafter, the resulting pulverized product was molded into a pellet having a diameter of 10 mm and a thickness of 5 mm and the pellet obtained was sintered in air atmosphere at 925° C. for 2 hours, whereby the dielectric ceramics were obtained.

The thus-obtained dielectric ceramics were processed to a size of 8 mm in diameter and 4 mm in thickness and then measured by a dielectric resonance method to calculate the $Q\times f^0$ value at the resonance frequency of 9 to 13 GHz, the dielectric constant ϵ_r and the temperature coefficient τ_f of resonance frequency. The results thereof are shown in Table 8.

Also, to 100 g of the dry-mixed powder obtained by mixing the main component, Li_2O powder, and the glass powder followed by solvent removal, 9 g of polyvinyl butyral as a binder, 6 g of dibutylphthalate as a plasticizer, and 60 g of toluene and 30 g of isopropyl alcohol both as a solvent were added to produce a green sheet having a thickness of 100 μm by the doctor blade method. Then, 20 layers of the green sheets were laminated by the thermo compression bonding of applying a pressure of 200 kg/cm^2 at a temperature of 65° C. At this time, a layer having been printed with Ag pattern as an internal electrode was disposed such that it was provided at the center in the thickness direction. After sintering the obtained laminated product at 925° C. for 2 hours, the sintered body was processed to a size of 5.0 mm in width, 1.5 mm in height and 9.5 mm in length, and an external electrode was formed to produce a tri-plate type resonator, as shown in FIGS. **1** and **2**. The obtained tri-plate type resonator was evaluated on the unloaded Q-value at a resonance frequency of 2.5 GHz. The result thereof is shown in Table 8.

Examples 64 to 75

In the same manner as in Example 63, the powder of the main component, Li_2O powder, and glass powder were weighed to have the composition ratios shown in Table 7, respectively. The weighed powders were then mixed and molded under the same condition as Example 63. The pellets obtained were sintered in air atmosphere at from 850 to 925° C. for 2 hours as shown in Table 7, whereby dielectric ceramics and resonators were obtained. The obtained dielectric ceramics and resonators were evaluated on various properties in the same method as in Example 63. The results thereof are shown in Table 8. FIG. **7** shows the X-ray diffraction pattern of the dielectric ceramics of Example 68, which is obtained by sintering the dielectric ceramic composition obtained by mixing the four-phase main component represented by aMg_2SiO_4-$bZnAl_2O_4$-$cSiO_2$-$dCaTiO_3$ according to the present invention, Li_2O as a subcomponent, and glass component.

Examples 76 to 84

ZnO and SiO_2 were weighed so that the molar ratio between them became 2:1. The weighed powders were charged into a ball mill together with ethanol and ZrO_2 ball and wet-mixed for 24 hours. After removing the solvent from the solution, the resulting mixture was dried and calcined at 1100° C. for 2 hours. The obtained calcined body is then pulverized to obtain a powder of Zn_2SiO_4 serving as the material of the main component. After that, in the same manner as in Example 63, the powder of the main component, Li_2O powder, and glass powder were weighed to have the composition ratios shown in Table 7, respectively. The weighed powders were then mixed and molded under the same condition as Example 63. The pellets obtained were sintered in air atmosphere at from 800 to 900° C. for 2 hours as shown in Table 8, whereby dielectric ceramics and resonators were obtained. The obtained dielectric ceramics and resonators were evaluated on various properties in the same method as in Example 63. The results thereof are shown in Table 8. FIG. **8** shows the X-ray diffraction pattern of the dielectric ceramics of Example 76, which is obtained by sintering the dielectric ceramic composition obtained by mixing the five-phase main component represented by aMg_2SiO_4-$bZnAl_2O_4$-$cSiO_2$-$dCaTiO_3$-eZn_2SiO_4 according to the present invention, Li_2O as a subcomponent, and glass component.

Comparative Examples 42 to 49

In the same manner as in Example 63, the calcined powder of the main component, Li_2O powder, and glass powder were weighed to have the composition ratios shown in Table 7, respectively. The weighed powders were then mixed and molded under the same condition as Example 63. The pellets obtained were sintered in air atmosphere at from 850 to 1000° C. for 2 hours as shown in Table 8, whereby dielectric ceramics and resonators were obtained (preferable sintered bodies were not obtained in some comparative examples). The obtained dielectric ceramics and resonators were evaluated on various properties in the same method as in Example 63. The results thereof are shown in Table 8.

Comparative Examples 50 to 54

In the same manner as in Example 76, the calcined powder of the main component, Li_2O powder, and glass powder were weighed to have the composition ratios shown in Table 7, respectively. The weighed powders were then mixed and molded under the same condition as Example 63. The pellets obtained were sintered in air atmosphere at from 825 to 1000° C. for 2 hours as shown in Table 8, whereby dielectric ceramics and resonators were obtained (preferable sintered bodies were not obtained in some comparative examples). The obtained dielectric ceramics and resonators were evaluated on various properties in the same method as in Example 63. The results thereof are shown in Table 8.

TABLE 1

	Main component composition(mol %)									
	ZnAl2O4	Zn2SiO4	TiO2	Zn2TiO4	Glass component composition(wt %)					
	a	b	c	d	SiO2	Al2O3	ZnO	PbO	Bi2O3	BaO
Example 1	7.50	67.50	25.00	0.00	6.0	11.0	47.0	—	—	4.0
Example 2	45.00	45.00	10.00	0.00	6.0	11.0	47.0	—	—	4.0
Example 3	45.00	45.00	10.00	0.00	26.0	1.0	12.0	30.0	1.0	—
Example 4	45.00	45.00	10.00	0.00	65.0	3.0	—	—	—	0.1
Example 5	60.00	32.50	7.50	0.00	6.0	11.0	47.0	—	—	4.0
Example 6	77.50	7.50	15.00	0.00	6.0	11.0	47.0	—	—	4.0
Example 7	80.00	10.00	10.00	0.00	6.0	11.0	47.0	—	—	4.0
Example 8	45.00	45.00	10.00	0.00	6.0	11.0	47.0	—	—	4.0
Example 9	45.00	45.00	10.00	0.00	6.0	11.0	47.0	—	—	4.0
Example 10	45.00	45.00	10.00	0.00	6.0	11.0	47.0	—	—	4.0
Example 11	7.50	52.50	20.00	20.00	6.0	11.0	47.0	—	—	4.0
Example 12	10.00	67.50	15.00	7.50	6.0	11.0	47.0	—	—	4.0
Example 13	30.00	30.00	15.00	25.00	6.0	11.0	47.0	—	—	4.0
Example 14	30.00	40.00	25.00	5.00	6.0	11.0	47.0	—	—	4.0
Example 15	44.00	40.00	15.00	1.00	6.0	11.0	47.0	—	—	4.0
Example 16	44.00	40.00	15.00	1.00	26.0	1.0	12.0	30.0	1.0	—
Example 17	44.00	40.00	15.00	1.00	65.0	3.0	—	—	—	0.1
Example 18	77.50	7.50	7.50	7.50	6.0	11.0	47.0	—	—	4.0
Example 19	80.00	9.00	10.90	0.10	6.0	11.0	47.0	—	—	4.0
Example 20	44.00	40.00	15.00	1.00	6.0	11.0	47.0	—	—	4.0
Example 21	44.00	40.00	15.00	1.00	6.0	11.0	47.0	—	—	4.0
Example 22	44.00	40.00	15.00	1.00	6.0	11.0	47.0	—	—	4.0
Comparative Example 1	2.50	70.00	27.50	0.00	6.0	11.0	47.0	—	—	4.0
Comparative Example 2	17.50	75.00	7.50	0.00	6.0	11.0	47.0	—	—	4.0
Comparative Example 3	50.00	47.50	2.50	0.00	6.0	11.0	47.0	—	—	4.0
Comparative Example 4	50.00	47.50	2.50	0.00	26.0	1.0	12.0	30.0	1.0	—
Comparative Example 5	50.00	47.50	2.50	0.00	65.0	3.0	—	—	—	0.1
Comparative Example 6	65.00	5.00	30.00	0.00	6.0	11.0	47.0	—	—	4.0
Comparative Example 7	77.50	2.50	20.00	0.00	6.0	11.0	47.0	—	—	4.0
Comparative Example 8	87.50	5.00	7.50	0.00	6.0	11.0	47.0	—	—	4.0
Comparative Example 9	2.50	60.00	25.00	12.50	6.0	11.0	47.0	—	—	4.0
Comparative Example 10	15.00	75.00	5.00	5.00	6.0	11.0	47.0	—	—	4.0
Comparative Example 11	25.00	42.50	30.00	2.50	6.0	11.0	47.0	—	—	4.0

TABLE 1-continued

Comparative Example 12	32.50	20.00	15.00	32.50	6.0	11.0	47.0	—	—	4.0
Comparative Example 13	40.00	20.00	30.00	10.00	6.0	11.0	47.0	—	—	4.0
Comparative Example 14	50.00	35.00	2.50	12.50	6.0	11.0	47.0	—	—	4.0
Comparative Example 15	70.00	2.50	15.00	12.50	6.0	11.0	47.0	—	—	4.0
Comparative Example 16	87.50	5.00	7.00	0.50	6.0	11.0	47.0	—	—	4.0
Comparative Example 17	44.00	40.00	15.00	1.00	6.0	11.0	47.0	—	—	4.0
Comparative Example 18	44.00	40.00	15.00	1.00	6.0	11.0	47.0	—	—	4.0

	Glass component composition(wt %)					mixing ratio between main component and glass component	
	SrO	CaO	SnO2	ZrO2	B2O3	Main component (part by weight)	Glass component (part by weight)
Example 1	0.2	0.8	1.0	—	30.0	100	30
Example 2	0.2	0.8	1.0	—	30.0	100	30
Example 3	—	—	—	—	30.0	100	30
Example 4	—	0.5	—	2.5	30.0	100	30
Example 5	0.2	0.8	1.0	—	30.0	100	30
Example 6	0.2	0.8	1.0	—	30.0	100	30
Example 7	0.2	0.8	1.0	—	30.0	100	30
Example 8	0.2	0.8	1.0	—	30.0	100	10
Example 9	0.2	0.8	1.0	—	30.0	100	70
Example 10	0.2	0.8	1.0	—	30.0	100	130
Example 11	0.2	0.8	1.0	—	30.0	100	30
Example 12	0.2	0.8	1.0	—	30.0	100	30
Example 13	0.2	0.8	1.0	—	30.0	100	30
Example 14	0.2	0.8	1.0	—	30.0	100	30
Example 15	0.2	0.8	1.0	—	30.0	100	30
Example 16	—	—	—	—	30.0	100	30
Example 17	—	0.5	—	2.5	30.0	100	30
Example 18	0.2	0.8	1.0	—	30.0	100	30
Example 19	0.2	0.8	1.0	—	30.0	100	30
Example 20	0.2	0.8	1.0	—	30.0	100	10
Example 21	0.2	0.8	1.0	—	30.0	100	70
Example 22	0.2	0.8	1.0	—	30.0	100	130
Comparative Example 1	0.2	0.8	1.0	—	30.0	100	30
Comparative Example 2	0.2	0.8	1.0	—	30.0	100	30
Comparative Example 3	0.2	0.8	1.0	—	30.0	100	30
Comparative Example 4	—	—	—	—	30.0	100	30
Comparative Example 5	—	0.5	—	2.5	30.0	100	30
Comparative Example 6	0.2	0.8	1.0	—	30.0	100	30
Comparative Example 7	0.2	0.8	1.0	—	30.0	100	30
Comparative Example 8	0.2	0.8	1.0	—	30.0	100	30
Comparative Example 9	0.2	0.8	1.0	—	30.0	100	30
Comparative Example 10	0.2	0.8	1.0	—	30.0	100	30
Comparative Example 11	0.2	0.8	1.0	—	30.0	100	30
Comparative Example 12	0.2	0.8	1.0	—	30.0	100	30
Comparative Example 13	0.2	0.8	1.0	—	30.0	100	30
Comparative Example 14	0.2	0.8	1.0	—	30.0	100	30
Comparative Example 15	0.2	0.8	1.0	—	30.0	100	30

TABLE 1-continued

Comparative Example 16	0.2	0.8	1.0	—	30.0	100	30
Comparative Example 17	0.2	0.8	1.0	—	30.0	100	3
Comparative Example 18	0.2	0.8	1.0	—	30.0	100	160

TABLE 2

	Sintering	Dielectric resonator charactristics			Tri-plate
	temperature (° C.)	εr (—)	Qxf0 (GHz)	τf (ppm/° C.)	Resonator Unloaded Q
Example 1	925	10.0	19620	18.1	185
Example 2	915	9.2	23190	−11.8	190
Example 3	905	9.7	16550	−14.1	195
Example 4	910	8.9	18960	−12.9	190
Example 5	915	9.0	23060	−15.4	195
Example 6	925	9.9	25650	−2.7	185
Example 7	925	9.4	25530	−11.1	185
Example 8	925	9.4	35440	−8.4	185
Example 9	905	8.9	16030	−15.5	195
Example 10	900	8.7	12700	−18.9	200
Example 11	875	10.0	11930	4.7	200
Example 12	875	9.5	15800	−3.2	200
Example 13	850	10.0	11300	−3.4	200
Example 14	850	10.0	17750	17.7	200
Example 15	865	9.5	21110	−1.0	205
Example 16	850	10.0	15460	−3.3	200
Example 17	875	9.2	17550	−2.1	200
Example 18	900	9.8	15050	−10.4	200
Example 19	900	9.8	22470	7.6	205
Example 20	905	9.7	30060	4.3	200
Example 21	850	8.7	15240	−7.8	200
Example 22	825	8.2	12330	−16.4	200
Comparative Example 1	925	10.2	5900	17.9	160
Comparative Example 2	915	8.0	19230	−34.0	195
Comparative Example 3	915	8.5	21950	−27.0	195
Comparative Example 4	905	9.0	15910	−29.5	195
Comparative Example 5	915	8.2	18130	−28.3	195
Comparative Example 6	925	11.7	25680	28.2	185
Comparative Example 7	Not sintered at 1000° C. or less				
Comparative Example 8	Not sintered at 1000° C. or less				
Comparative Example 9	875	10.7	4230	14.4	155
Comparative Example 10	875	8.0	18200	−39.0	205
Comparative Example 11	850	11.0	18990	28.3	205
Comparative Example 12	850	11.2	3200	−4.4	150
Comparative Example 13	875	11.9	15620	29.4	200
Comparative Example 14	895	9.1	15370	−25.0	200
Comparative Example 15	Not sintered at 1000° C. or less				
Comparative Example 16	Not sintered at 1000° C. or less				
Comparative Example 17	Not sintered at 1000° C. or less				
Comparative Example 18	Glass was eluted				

TABLE 3

	Main component composition (mol %)							Glass component composition (wt %)	
	a ZnO	b Al2O3	c SiO2	d CaO + TiO2	x CaO/ (CaO + TiO2)	1 − x TiO2/ (CaO + TiO2)	Subcomponent Li2O (part by		
	a	b	c	d	e	e	weight)	SiO2	Al2O3
Example 23	10.0	35.0	35.0	20.0	0.0	1.0	5	6.0	12.0
Example 24	30.0	25.0	25.0	20.0	0.0	1.0	5	6.0	12.0
Example 25	40.0	25.0	25.0	10.0	0.0	1.0	5	6.0	12.0
Example 26	40.0	25.0	25.0	10.0	0.3	0.8	5	6.0	12.0
Example 27	40.0	25.0	25.0	10.0	0.7	0.3	5	6.0	12.0
Example 28	10.0	10.0	65.0	15.0	0.5	0.5	5	6.0	12.0
Example 29	10.0	35.0	35.0	20.0	0.5	0.5	5	6.0	12.0
Example 30	10.0	60.0	10.0	20.0	0.5	0.5	5	6.0	12.0
Example 31	25.0	25.0	25.0	25.0	0.5	0.5	5	6.0	12.0
Example 32	50.0	15.0	15.0	20.0	0.5	0.5	5	6.0	12.0
Example 33	40.0	20.0	20.0	20.0	0.5	0.5	5	6.0	12.0
Example 34	40.0	20.0	20.0	20.0	0.5	0.5	10	6.0	12.0
Example 35	40.0	20.0	20.0	20.0	0.5	0.5	25	6.0	12.0
Example 36	30.0	25.0	25.0	20.0	0.0	1.0	5	21.0	12.0
Example 37	30.0	25.0	25.0	20.0	0.0	1.0	5	6.0	12.0
Example 38	40.0	20.0	20.0	20.0	0.5	0.5	10	27.0	—
Example 39	40.0	20.0	20.0	20.0	0.5	0.5	10	67.0	2.2

TABLE 3-continued

Example 40	35.0	25.0	25.0	15.0	0.5	0.5	10	6.0	12.0
Example 41	35.0	25.0	25.0	15.0	0.5	0.5	10	6.0	12.0
Example 42	35.0	25.0	25.0	15.0	0.5	0.5	10	6.0	12.0
Comparative Example 19	5.0	35.0	30.0	15.0	0.5	0.5	5	6.0	12.0
Comparative Example 20	60.0	15.0	15.0	10.0	0.5	0.5	5	6.0	12.0
Comparative Example 21	10.0	70.0	2.5	17.5	0.5	0.5	5	6.0	12.0
Comparative Example 22	12.5	2.5	75.0	10.0	0.5	0.5	5	6.0	12.0
Comparative Example 23	40.0	25.0	25.0	10.0	0.9	1.0	5	6.0	12.0
Comparative Example 24	45.0	25.0	25.0	5.0	0.5	0.5	5	6.0	12.0
Comparative Example 25	30.0	20.0	20.0	30.0	0.5	0.5	5	6.0	12.0
Comparative Example 26	30.0	25.0	25.0	20.0	0.5	0.5	0.5	6.0	12.0
Comparative Example 27	30.0	25.0	25.0	20.0	0.5	0.5	35	6.0	12.0
Comparative Example 28	35.0	25.0	25.0	15.0	0.7	0.3	10	6.0	12.0
Comparative Example 29	35.0	25.0	25.0	15.0	0.7	0.3	10	6.0	12.0

	Glass component composition (wt %)								Mixing ratio of main component and glass component	
	ZnO	PbO	Bi2O3	BaO	SrO	SnO2	ZrO2	B2O3	Main component (part by weight)	Glass component (part by weight)
Example 23	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Example 24	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Example 25	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Example 26	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Example 27	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Example 28	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Example 29	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Example 30	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Example 31	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Example 32	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Example 33	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Example 34	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Example 35	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Example 36	47.0	—	—	3.0	1.0	1.0	—	15.0	100	20
Example 37	32.0	—	—	3.0	1.0	1.0	—	45.0	100	20
Example 38	12.0	30.0	1.0	—	—	—	—	30.0	100	20
Example 39	—	—	—	0.1	—	—	0.7	30.0	100	20
Example 40	47.0	—	—	3.0	1.0	1.0	—	30.0	100	10
Example 41	47.0	—	—	3.0	1.0	1.0	—	30.0	100	50
Example 42	47.0	—	—	3.0	1.0	1.0	—	30.0	100	130
Comparative Example 19	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Comparative Example 20	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Comparative Example 21	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Comparative Example 22	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Comparative Example 23	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Comparative Example 24	47.0	—	—	3.0	1.0	1.0	—	30.0	100	40
Comparative Example 25	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Comparative Example 26	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Comparative Example 27	47.0	—	—	3.0	1.0	1.0	—	30.0	100	20
Comparative Example 28	47.0	—	—	3.0	1.0	1.0	—	30.0	100	2.5
Comparative Example 29	47.0	—	—	3.0	1.0	1.0	—	30.0	100	160

TABLE 4

	Sintering	Dielectric resonator characteristics			Tri-plate
	temperature (° C.)	εr (—)	Qxf0 (GHz)	τf (ppm/° C.)	resonator Unloaded Q
Example 23	975	8.8	21000	10.0	235
Example 24	975	9.8	23150	5.0	240
Example 25	975	9.1	26220	−18.4	250
Example 26	925	9.3	24870	−10.9	245
Example 27	875	9.7	23050	−8.5	240
Example 28	875	7.5	27250	7.4	255
Example 29	900	8.2	23000	13.7	240
Example 30	900	9.6	22100	0.0	240
Example 31	875	9.8	21130	18.1	235
Example 32	900	9.6	20920	0.5	235
Example 33	875	9.5	22300	1.8	240
Example 34	875	9.1	23270	−1.5	240
Example 35	875	8.7	20960	−16.3	235
Example 36	975	9.5	23950	3.9	245
Example 37	950	9.8	21150	4.9	235
Example 38	900	9.7	22240	−2.7	240
Example 39	900	8.5	23820	−3.4	245
Example 40	925	9.7	28780	−1.0	260
Example 41	875	9.2	23920	−7.9	245
Example 42	850	8.8	20110	−16.6	230
Comparative Example 19		Not sintered at 1000° C. or less			
Comparative Example 20	900	9.4	4300	−12.3	150

TABLE 4-continued

	Sintering	Dielectric resonator characteristics			Tri-plate
	temperature (° C.)	εr (—)	Qxf0 (GHz)	τf (ppm/° C.)	resonator Unloaded Q
Comparative Example 21		Not sintered at 1000° C. or less			
Comparative Example 22	900	7.2	5200	−21.0	165
Comparative Example 23	925	12.6	8700	42.9	180
Comparative Example 24	900	8.6	13000	−34.0	180
Comparative Example 25	875	8.2	12500	−40.0	180
Comparative Example 26		Not sintered at 1000° C. or less			
Comparative Example 27		Glass was eluted			
Comparative Example 28		Not sintered at 1000° C. or less			
Comparative Example 29		Glass was eluted			

TABLE 5

	Main component composition (mol %)				Subcomponent					
	a ZnAl2O4	b Zn2SiO4	c SiO2	d SrTiO3	Li2CO3 (part by	Glass component composition(wt %)				
	a	b	c	d	weight)	SiO2	Al2O3	ZnO	PbO	Bi2O3
Example 43	75.0	5.0	5.0	15.0	5	6.0	12.0	47.0	—	—
Example 44	55.0	25.0	5.0	15.0	5	6.0	12.0	47.0	—	—
Example 45	40.0	40.0	5.0	15.0	5	6.0	12.0	47.0	—	—
Example 46	25.0	55.0	5.0	15.0	5	6.0	12.0	47.0	—	—
Example 47	5.0	75.0	5.0	15.0	5	6.0	12.0	47.0	—	—
Example 48	35.0	35.0	10.0	15.0	5	6.0	12.0	47.0	—	—
Example 49	32.5	32.5	20.0	15.0	5	6.0	12.0	47.0	—	—
Example 50	27.5	27.5	30.0	15.0	5	6.0	12.0	47.0	—	—
Example 51	25.0	25.0	35.0	15.0	5	6.0	12.0	47.0	—	—
Example 52	40.0	32.5	12.5	12.5	5	6.0	12.0	47.0	—	—
Example 53	35.0	22.5	30.0	12.5	5	6.0	12.0	47.0	—	—
Example 54	40.0	30.0	15.0	15.0	10	6.0	12.0	47.0	—	—
Example 55	40.0	30.0	15.0	15.0	25	6.0	12.0	47.0	—	—
Example 56	35.0	22.5	30.0	12.5	5	21.0	12.0	47.0	—	—
Example 57	35.0	22.5	30.0	12.5	5	6.0	12.0	32.0	—	—
Example 58	40.0	30.0	15.0	15.0	10	27.0	—	12.0	30.0	1.0
Example 59	40.0	30.0	15.0	15.0	10	67.0	2.2	—	—	—
Example 60	40.0	32.5	12.5	12.5	10	6.0	12.0	47.0	—	—
Example 61	40.0	32.5	12.5	12.5	10	6.0	12.0	47.0	—	—
Example 62	40.0	32.5	12.5	12.5	10	6.0	12.0	47.0	—	—

TABLE 5-continued

Comparative Example 30	80.0	2.5	5.0	12.5	5	6.0	12.0	47.0	—	—
Comparative Example 31	1.0	54.0	30.0	15.0	5	6.0	12.0	47.0	—	—
Comparative Example 32	2.5	80.0	5.0	12.5	5	6.0	12.0	47.0	—	—
Comparative Example 33	54.0	1.0	30.0	15.0	5	6.0	12.0	47.0	—	—
Comparative Example 34	42.0	42.0	1.0	15.0	5	6.0	12.0	47.0	—	—
Comparative Example 35	25.0	17.5	40.0	17.5	5	6.0	12.0	47.0	—	—
Comparative Example 36	35.0	27.5	30.0	7.5	5	6.0	12.0	47.0	—	—
Comparative Example 37	25.0	20.0	35.0	20.0	5	6.0	12.0	47.0	—	—
Comparative Example 38	40.0	32.5	12.5	12.5	0.5	6.0	12.0	47.0	—	—
Comparative Example 39	40.0	32.5	12.5	12.5	35	6.0	12.0	47.0	—	—
Comparative Example 40	40.0	32.5	12.5	12.5	10	6.0	12.0	47.0	—	—
Comparative Example 41	40.0	32.5	12.5	12.5	10	6.0	12.0	47.0	—	—

	Glass component composition(wt %)					Mixing ratio of main component and glass component	
	BaO	SrO	SnO2	ZrO2	B2O3	Main component (part by weight)	Glass component (part by weight)
Example 43	3.0	1.0	1.0	—	30	100	25
Example 44	3.0	1.0	1.0	—	30	100	25
Example 45	3.0	1.0	1.0	—	30	100	25
Example 46	3.0	1.0	1.0	—	30	100	25
Example 47	3.0	1.0	1.0	—	30	100	25
Example 48	3.0	1.0	1.0	—	30	100	25
Example 49	3.0	1.0	1.0	—	30	100	25
Example 50	3.0	1.0	1.0	—	30	100	25
Example 51	3.0	1.0	1.0	—	30	100	25
Example 52	3.0	1.0	1.0	—	30	100	25
Example 53	3.0	1.0	1.0	—	30	100	25
Example 54	3.0	1.0	1.0	—	30	100	25
Example 55	3.0	1.0	1.0	—	30	100	25
Example 56	3.0	1.0	1.0	—	15	100	25
Example 57	3.0	1.0	1.0	—	45	100	25
Example 58	—	—	—	—	30	100	25
Example 59	0.1	—	—	0.7	30	100	25
Example 60	3.0	1.0	1.0	—	30	100	10
Example 61	3.0	1.0	1.0	—	30	100	50
Example 62	3.0	1.0	1.0	—	30	100	130
Comparative Example 30	3.0	1.0	1.0	—	30	100	130
Comparative Example 31	3.0	1.0	1.0	—	30	100	25
Comparative Example 32	3.0	1.0	1.0	—	30	100	25
Comparative Example 33	3.0	1.0	1.0	—	30	100	130
Comparative Example 34	3.0	1.0	1.0	—	30	100	130
Comparative Example 35	3.0	1.0	1.0	—	30	100	25
Comparative Example 36	3.0	1.0	1.0	—	30	100	25
Comparative Example 37	3.0	1.0	1.0	—	30	100	25
Comparative Example 38	3.0	1.0	1.0	—	30	100	130
Comparative Example 39	3.0	1.0	1.0	—	30	100	10

TABLE 5-continued

Comparative Example 40	3.0	1.0	1.0	—	30	100	2.5
Comparative Example 41	3.0	1.0	1.0	—	30	100	160

TABLE 6

	Sintering	Dielectric resonator characteristics			Tri-plate
	temperature (° C.)	εr (—)	QxfO (GHz)	τf (ppm/° C.)	resonator Unloaded Q
Example 43	975	9.8	25420	4.7	250
Example 44	925	9.5	24070	1.7	245
Example 45	900	9.4	23130	−2.1	240
Example 46	900	9.3	22270	−6.3	240
Example 47	875	9.1	21260	−18.0	235
Example 48	900	9.5	21860	7.8	240
Example 49	925	9.3	21640	6.4	235
Example 50	950	9.3	20590	14.1	230
Example 51	950	9.2	20050	18.0	230
Example 52	900	9.4	22620	2.5	240
Example 53	925	8.8	23600	−12.0	240
Example 54	875	9.4	22440	4.0	240
Example 55	850	9.0	20520	−10.0	230
Example 56	950	8.4	24110	−7.3	245
Example 57	900	8.7	21380	−4.9	235
Example 58	875	9.7	22000	2.8	240
Example 59	925	8.7	22730	2.4	240
Example 60	975	9.7	28340	5.5	260
Example 61	875	9.2	24320	0.9	245
Example 62	825	8.8	20810	−7.8	230
Comparative Example 30	Not sintered at 1000° C. or less				
Comparative Example 31	825	8.8	6000	−36.7	165
Comparative Example 32	825	8.7	8360	−33.0	170
Comparative Example 33	Not sintered at 1000° C. or less				
Comparative Example 34	Not sintered at 1000° C. or less				
Comparative Example 35	825	9.5	9960	32.0	175
Comparative Example 36	825	7.9	13300	−62.7	180
Comparative Example 37	900	10.5	12520	80.7	180
Comparative Example 38	Not sintered at 1000° C. or less				
Comparative Example 39	Glass was eluted				
Comparative Example 40	Not sintered at 1000° C. or less				
Comparative Example 41	Glass was eluted				

TABLE 7

	Main component (molar fraction)					subcomponent Li2O	Glass component				
	a	b	c	d	e	(part by	composition (wt %)				
	Mg2SiO4	ZnAl2O4	SiO2	CaTiO3	Zn2SiO4	weight)	SiO2	Al2O3	ZnO	PbO	Bi2O3
Example 63	0.12	0.58	0.10	0.20	0	5	6.0	12.0	25.0	—	—
Example 64	0.35	0.35	0.10	0.20	0	5	6.0	12.0	25.0	—	—
Example 65	0.70	0.10	0.05	0.15	0	5	6.0	12.0	25.0	—	—
Example 66	0.30	0.30	0.20	0.20	0	5	6.0	12.0	25.0	—	—
Example 67	0.15	0.55	0.15	0.15	0	5	6.0	12.0	25.0	—	—
Example 68	0.40	0.25	0.15	0.20	0	10	6.0	12.0	25.0	—	—
Example 69	0.40	0.25	0.15	0.20	0	10	21.0	12.0	47.0	—	—
Example 70	0.40	0.25	0.15	0.20	0	10	6.0	12.0	32.0	—	—
Example 71	0.40	0.25	0.15	0.20	0	5	27.0	—	12.0	30.0	1.0

TABLE 7-continued

Example 72	0.40	0.25	0.15	0.20	0	5	67.0	2.2	—	—	—
Example 73	0.43	0.25	0.17	0.15	0	10	6.0	12.0	25.0	—	—
Example 74	0.43	0.25	0.17	0.15	0	10	6.0	12.0	25.0	—	—
Example 75	0.43	0.25	0.17	0.15	0	10	6.0	12.0	25.0	—	—
Example 76	0.45	0.20	0.12	0.20	0.03	5	6.0	12.0	25.0	—	—
Example 77	0.28	0.28	0.19	0.20	0.05	5	6.0	12.0	25.0	—	—
Example 78	0.15	0.50	0.18	0.15	0.02	10	6.0	12.0	25.0	—	—
Example 79	0.30	0.30	0.15	0.20	0.05	5	21.0	12.0	47.0	—	—
Example 80	0.30	0.30	0.15	0.20	0.05	5	6.0	12.0	32.0	—	—
Example 81	0.30	0.30	0.15	0.20	0.05	5	27.0	—	12.0	30.0	1.0
Example 82	0.30	0.30	0.15	0.20	0.05	10	67.0	2.2	—	—	—
Example 83	0.28	0.28	0.19	0.20	0.05	10	6.0	12.0	25.0	—	—
Example 84	0.15	0.50	0.17	0.15	0.03	5	6.0	12.0	25.0	—	—
Comparative Example 42	0.75	0.07	0.05	0.13	0.00	10	6.0	12.0	25.0	—	—
Comparative Example 43	0.02	0.60	0.15	0.20	0.00	10	6.0	12.0	25.0	—	—
Comparative Example 44	0.10	0.65	0.10	0.15	0.00	10	6.0	12.0	25.0	—	—
Comparative Example 45	0.60	0.05	0.15	0.20	0.00	5	6.0	12.0	25.0	—	—
Comparative Example 46	0.40	0.40	0.01	0.19	0.00	5	6.0	12.0	25.0	—	—
Comparative Example 47	0.26	0.26	0.28	0.20	0.00	5	6.0	12.0	25.0	—	—
Comparative Example 48	0.30	0.40	0.20	0.10	0.00	5	6.0	12.0	25.0	—	—
Comparative Example 49	0.30	0.30	0.15	0.25	0.00	5	6.0	12.0	25.0	—	—
Comparative Example 50	0.30	0.30	0.10	0.20	0.10	5	6.0	12.0	25.0	—	—
Comparative Example 51	0.35	0.35	0.10	0.15	0.05	0.5	6.0	12.0	25.0	—	—
Comparative Example 52	0.35	0.30	0.17	0.15	0.03	20	6.0	12.0	25.0	—	—
Comparative Example 53	0.35	0.30	0.17	0.15	0.03	10	6.0	12.0	25.0	—	—
Comparative Example 54	0.30	0.30	0.15	0.20	0.05	5	6.0	12.0	25.0	—	—

	Glass component composition (wt %)					Mixing ratio of main component and glass component	
	BaO	SrO	SnO2	ZrO2	B2O3	Main component (part by weight)	Glass component (part by weight)
Example 63	25.0	1.0	1.0	—	30.0	100	25
Example 64	25.0	1.0	1.0	—	30.0	100	25
Example 65	25.0	1.0	1.0	—	30.0	100	25
Example 66	25.0	1.0	1.0	—	30.0	100	25
Example 67	25.0	1.0	1.0	—	30.0	100	25
Example 68	25.0	1.0	1.0	—	30.0	100	25
Example 69	3.0	1.0	1.0	—	15.0	100	25
Example 70	3.0	1.0	1.0	—	45.0	100	25
Example 71	—	—	—	—	30.0	100	25
Example 72	0.1	—	—	0.7	30.0	100	25
Example 73	25.0	1.0	1.0	—	30.0	100	10
Example 74	25.0	1.0	1.0	—	30.0	100	70
Example 75	25.0	1.0	1.0	—	30.0	100	130
Example 76	25.0	1.0	1.0	—	30.0	100	25
Example 77	25.0	1.0	1.0	—	30.0	100	25
Example 78	25.0	1.0	1.0	—	30.0	100	25
Example 79	3.0	1.0	1.0	—	15.0	100	25
Example 80	3.0	1.0	1.0	—	45.0	100	25
Example 81	—	—	—	—	30.0	100	50
Example 82	0.1	—	—	0.7	30.0	100	25
Example 83	25.0	1.0	1.0	—	30.0	100	25
Example 84	25.0	1.0	1.0	—	30.0	100	130
Comparative Example 42	25.0	1.0	1.0	—	30.0	100	25
Comparative Example 43	25.0	1.0	1.0	—	30.0	100	25
Comparative Example 44	25.0	1.0	1.0	—	30.0	100	25

TABLE 7-continued

Comparative Example 45	25.0	1.0	1.0	—	30.0	100	25
Comparative Example 46	25.0	1.0	1.0	—	30.0	100	25
Comparative Example 47	25.0	1.0	1.0	—	30.0	100	25
Comparative Example 48	25.0	1.0	1.0	—	30.0	100	25
Comparative Example 49	25.0	1.0	1.0	—	30.0	100	25
Comparative Example 50	25.0	1.0	1.0	—	30.0	100	25
Comparative Example 51	25.0	1.0	1.0	—	30.0	100	25
Comparative Example 52	25.0	1.0	1.0	—	30.0	100	25
Comparative Example 53	25.0	1.0	1.0	—	30.0	100	2.5
Comparative Example 54	25.0	1.0	1.0	—	30.0	100	160

TABLE 8

	Sintering	Dielectric resonator characteristics			Tri-plate
	temperature (° C.)	εr (—)	QxfO (GHz)	τf (ppm/° C.)	resonator Unloaded Q
Example 63	925	7.8	28100	−16.0	275
Example 64	900	9.5	25800	−14.5	265
Example 65	875	9.8	20300	−17.3	230
Example 66	900	8.3	24500	−14.7	245
Example 67	925	9.9	28300	−18.4	275
Example 68	875	9.4	23700	−14.8	240
Example 69	925	9.0	25300	−12.3	250
Example 70	875	9.2	22000	−8.1	240
Example 71	850	9.7	20300	−11.2	230
Example 72	925	8.7	20700	−12.7	235
Example 73	925	9.7	25400	−8.8	250
Example 74	875	9.2	22100	−16.2	240
Example 75	850	8.4	21600	−18.6	240
Example 76	825	9.7	20300	−16.8	230
Example 77	825	9.4	21300	−18.5	240
Example 78	875	8.8	23500	−17.5	245
Example 79	875	9.3	26700	−14.1	270
Example 80	850	9.5	23200	−12.9	240
Example 81	850	9.4	21000	−14.5	235
Example 82	900	9.8	24400	−10.8	245
Example 83	875	9.4	23600	−14.2	245
Example 84	800	8.3	20400	−17.9	240
Comparative Example 42	850	10.8	8320	−18.5	175
Comparative Example 43	Not sintered at 1000° C. or less				
Comparative Example 44	Not sintered at 1000° C. or less				
Comparative Example 45	875	10.5	7200	−15.0	170
Comparative Example 46	Not sintered at 1000° C. or less				
Comparative Example 47	900	7.6	3500	−17.8	150
Comparative Example 48	875	8.2	8530	−39.7	175
Comparative Example 49	900	13.2	5200	53.0	160
Comparative Example 50	825	10.8	6700	−17.0	165
Comparative Example 51	Not sintered at 1000° C. or less				
Comparative Example 52	Glass was eluted				
Comparative Example 53	Not sintered at 1000° C. or less				
Comparative Example 54	Glass was eluted				

INDUSTRIAL APPLICABILITY

According to the present invention, it is possible to provide a dielectric ceramic composition that can be sintered at a sintering temperature of 1000° C. or less and is sintered to form dielectric ceramics having a dielectric constant ϵ_r of not more than 10, a large Q value in high-frequency region, and an absolute value in temperature coefficient τ_f of resonance frequency of not more than 20 ppm/° C. Since the dielectric ceramic composition can be sintered at a sintering temperature of 1000° C. or less, it is possible to reduce an electric power required for the sintering, to perform simultaneous sintering with a low resistant conductor such as Cu or Ag at relatively low cost, and to provide a laminated ceramic part having an internal electrode comprising Ag or Cu.

What is claimed is:

1. A dielectric ceramic composition containing a Li compound as a subcomponent in an amount of 1 to 15 parts by weight in terms of Li_2O and a glass component in an amount of 5 to 150 parts by weight, based on 100 parts by weight of a main component represented by general formula (4): aMg_2SiO_4-$bZnAl_2O_4$-$cSiO_2$-$dCaTiO_3$-eZn_2SiO_4, in which the molar fractions of respective components a, b, c, d, and e satisfy $0.10 \leqq a \leqq 0.72$, $0.08 \leqq b \leqq 0.62$, $0.02c \leqq 0.22$, $0.12 \leqq d \leqq 0.22$, $0 \leqq e \leqq 0.08$ $(a+b+c+d+e=1)$.

2. The dielectric ceramic composition as claimed in claim **1**, wherein the glass component includes one or more glasses selected from a PbO-base glass, a ZnO-base glass, a SiO_2-base glass, a B_2O_3-base glass, and a glass comprising two or more oxides selected from the group consisting of SiO_2, Al_{23}, ZnO, PbO, Bi_2O_3, BaO, SrO, SnO_2, ZrO_2, and B_2O_3.

3. The dielectric ceramic composition as claimed in claim **2**, wherein the glass component is composed of SiO_2 in an amount of 2 to 70 wt %, Al_2O_3 in an amount of 0 to 15 wt %, ZnO in an amount of 10 to 55 wt %, PbO in an amount of 0 to 35 wt %, Bi_2O_3 in an amount of 0 to 2 wt %, BaO in an amount of 0 to 30 wt %, SrO in an amount of 0 to 2 wt %, SnO_2 in an amount of 0 to 2 wt %, ZrO_2 in an amount of 0 to 1 wt %, and B_2O_3 in an amount of 10 to 50wt %.

4. Dielectric ceramics containing crystalline phases of Mg_2SiO_4, $ZnAl_2O_4$, SiO_2, and $CaTiO_3$ and a glass phase, which is obtained by sintering the dielectric ceramic composition as claimed in claim **1**.

5. Dielectric ceramics containing crystalline phases of Mg_2SiO_4, $ZnAl_2O_4$, SiO_2, $CaTiO_3$, and Zn_2SiO_4 and a glass phase, which is obtained by sintering the dielectric ceramic composition as claimed in claim **1**.

6. A material composition for dielectric ceramics represented by general formula (4): aMg_2SiO_4-$bZnAl_2O_4$-$cSiO_2$-$dCaTiO_3$-eZn_2SiO_4, in which the molar fractions of respective components a, b, c, d, and e satisfy $0.10 \leqq a \leqq 0.72$, $0.08 \leqq b \leqq 0.62$, $0.02 \leqq c \leqq 0.22$, $012 \leqq d \leqq 0.22$, $0 \leqq e \leqq 0.08$ $(a+b+c+d+e=1$.

7. A laminated ceramic part having a plurality of dielectric layers, an internal electrode formed between the dielectric layers and an external electrode electrically connected to the internal electrode, wherein the dielectric layers are constituted of dielectric ceramics obtained by sintering the dielectric ceramic composition as claimed in claim **1**, and the internal electrode is made of elemental Cu or elemental Ag, or an alloy material mainly comprising Cu or Ag.

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